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CALCULATION OF ATMOSPHERIC COMPOSITION IN THE HIGH LATITUDE SEP--ETC(U)
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CALCULATION OF ATMOSPHERIC COMPOSITION IN THE HIGH LATITUDE SEPTEMBER STRATOSPHERE

MAY 1978

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Prediction of prenuclear event atmospheric composition in the 10 to 100 km region of the atmosphere is required for initial input to models of nuclear weapons effects. ANMAR (the ASL Numerical Model of Atmospheric Radiation, Composition and Dynamics) was ~~utilized~~ used to generate an atmospheric composition model of the 65 deg N September stratosphere (10 to 50 km). Model results of concentrations for $O(^3P)$, $O_2(^1\Delta)$, O_3, NO, NO_2, NO_3, N_2O_5, HNO_3, N_2O, HNO_2, $O(^1D)$, H_2O, OH, H_2O_2, HO_2, CH_4, HCHO, and CO are presented as number density and mixing — over

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20. ABSTRACT (cont)

ratios in graphical form. Resultant volume production rates for several important reactions are also presented. The calculations performed by the model are fully time dependent and diurnal; however, only the results for noontime are shown. Comparison of calculated results with some observations shows reasonable agreement.

CONTENTS

INTRODUCTION

In the areas of ballistic missile defense, communications, electronic warfare, and missile design and testing, the investigation of nuclear effects on electromagnetic wave propagation is necessary. Predictive models of atmospheric modification in the 10 to 100 km altitude region resulting from nuclear events require knowledge of prenuclear atmospheric composition. It is within the 10 to 100 km altitude interval that least information is available concerning atmospheric composition.

The existence of a functioning computer program to model atmospheric composition [1] makes it possible to provide theoretical support to ASL researchers in their ongoing efforts to study the conditions and composition of the atmosphere. A previous report [2] was released to present part of the results obtained for the 32° N September stratosphere. Hence, as a followup, the computerized model was recently utilized to generate comparative results for the 65° N September stratosphere.

The basic chemical reactions used are the same as partially described in [2] and more completely in [3]. Photoreaction coefficients were computed by J. L. Collins and are described in [4].

In attempting to deal with transport the same approach as discussed in [2] was used to develop a model of the chemical composition in the September stratosphere at 65° N latitude.

The required temperature profile was obtained from R. O. Olsen of the Atmospheric Sciences Laboratory. A literature search was performed by J. L. Collins, formerly of the University of Texas at El Paso, to obtain initial values of concentrations for some key species.

COMPUTATIONAL RESULTS

A similar format for presentation as was used to describe the results of the 32° N September stratosphere is followed to give the results for the 65° N September stratosphere.

Consequently, the categories of calculated results presented consist of: (1) particle densities of selected important species, (2) mixing ratios by volume of these same species, and (3) reaction rates of several sets of some key photolytic and chemical reactions. All results shown are given as a function of altitude between 10 and 50 km at noontime. Computations were done at 5 km intervals.

From the nitrogen/oxygen family, Figs. 1a through 1k are computed altitude profiles of the particle densities of ground-state oxygen atoms ($O(^3P)$), excited oxygen molecules ($O_2(^1\Delta)$), ozone (O_3), nitric oxide (NO), nitrogen dioxide (NO_2), NO_3 radical, dinitrogen pentoxide (N_2O_5), nitric acid (HNO_3), nitrous oxide (N_2O), along with nitrous acid (HNO_2), and electronically excited oxygen atoms $O(^1D)$.

From the oxygen/hydrogen family, Figs. 1ℓ through 1o are the computed particle densities for water (H_2O), hydroxyl radical (OH), hydrogen peroxide (H_2O_2) and perhydroxyl radical (HO_2).

Methane (CH_4), formaldehyde (HCHO), and carbon monoxide (CO) from the carbon/hydrogen/oxygen family of reactions are shown in Figs. 1p, 1q, and 1r.

Existence of some of the species in the atmosphere is speculative; but since their presence is required by photolytic and chemical considerations, these species are also presented here. For some species which have been detected in the atmosphere, the measured results are plotted along with the computed values to offer a comparison between theoretical calculations and experimental measurements.

Mixing ratios for the same data as 1a through 1r are presented in Figs. 2a through 2r to allow direct consideration of the fraction of the atmosphere that a given constituent represents. This ratio of the numbers of the particles is termed the volume mixing ratio and is computed by dividing the individual particle density by the total particle density of all the species present in the volume considered. These figures express the fact that species which play dominant roles in many atmospheric processes exist in only extremely small proportions in comparison to other less chemically important species.

To better understand the role of various chemical reactions, one should consider the rates at which given reactions occur under the prescribed conditions. Hence Figs. 3a through 3n are the calculated altitude dependent rates of a number of reactions for the 65° N September situation. Although these rates are labeled "reaction rates," they are more precisely defined "volume production rates."

A partial list of the reactions is shown in Table 2. On the graphs, some of the reactions are grouped to allow immediate observation of certain critical relationships between species. These relationships are described in [2].

THE ATMOSPHERIC CHEMICAL KINETICS MODEL

The design and calculational method used in the computer simulation of stratospheric composition as well as the structure of the 34 photodissociative processes and 115 chemical reactions are discussed in [2] and [3]. The atmospheric molecules, atoms, and free radicals used in the modeling effort are shown in Table 1. A selected list of reactions with rate constants is given in Table 2.

TABLE 1

ATMOSPHERIC CHEMICAL SPECIES CONSIDERED IN COMPUTATIONAL MODEL

O $O(^1D)$ $O(^1S)$ O_2 $O_2(^1\Delta)$ $O_2(^1\Sigma)$ O_3

N N_2 NO NO_2 N_2O NO_3 N_2O_5

H H_2 OH H_2O HO_2 H_2O_2

CO CO_2 CH_2 CH_3 CH_4 CHO $HCHO$

CH_3O CH_3O_2 CH_3OOH

HNO_2 HNO_3

COMPARISON OF MEASUREMENTS AND CALCULATIONS

Results of some experimental measurements at higher latitudes are plotted on the graphs of calculated particle densities and mixing ratios. In many cases referenced literature contained information on either mixing ratio or number density, and the other factor had to be computed from the available data. When computation was necessary, the total number density values were taken from tables in the US Standard Atmosphere of 1976.

For ozone, only the measurements by Randhawa [5] on two different days in September at Poker Flat are used. As can be seen in Figs. 1c and 2c, the computational results are very close to the experimentally measured values. Although calculated values for ozone at 32° N latitude [2] are not shown here, the calculated values at 65° N latitude were found to be generally higher than at 32° N latitude in the lower stratosphere and lower in the upper stratosphere (25 to 50 km).

Nitric oxide calculated values are shown in Figs. 1d and 2d with experimentally measured values by Ridley et al. [6] taken at 59° N in July 1974 and by Lowenstein and Savage [7] in June 1975. Comparison with Ridley's measurements show the calculated NO profile to be somewhat high. However, when calculated values of NO are compared with Lowenstein and Savage's measurements, agreement is quite good at the altitudes where those measurements were made.

In Figs. 1e and 2e calculated values of nitrogen dioxide are fairly close to measured values of Kerr and McElroy [8]. The measurements of August 17 were taken in the evening and those of August 18 were recorded in the morning. Comparison with the measurements of Evans et al. [9] shows good agreement except for a shift in the region of maximum values and possibly a sharper decrease in NO_2 concentration above 30 km.

TABLE 2. REACTIONS SELECTED TO ILLUSTRATE ALTITUDE-DEPENDENCE OF RATES FOR FIGURES OF SECTION 3

				Rate Constants*								
				10 km	15 km	20 km	25 km	30 km	35 km	40 km	45 km	50 km
(1)	O_2 + hν	→	O + O	4.9(-20)	1.4(-16)	5.6(-14)	2.9(-12)	2.0(-11)	1.0(-10)	4.6(-10)	8.0(-10)	1.1(-9)
(5)	O_3 + hν	→	O + O_2	2.7(-4)	2.8(-4)	2.9(-4)	2.9(-4)	2.9(-4)	3.0(-4)	3.0(-4)	3.1(-4)	3.1(-4)
(7)	O_3 + hν	→	$O(^1D) + O_2(^1\Delta)$	2.3(-6)	5.0(-6)	1.2(-5)	3.6(-5)	1.6(-4)	4.7(-4)	1.1(-3)	2.5(-3)	3.8(-3)
(11)	NO_2 + hν	→	O + NO	1.0(-2)	1.0(-2)	1.1(-2)	1.2(-2)	1.2(-2)	1.3(-2)	1.3(-2)	1.3(-2)	1.3(-2)
(32)	HNO_3 + hν	→	OH + NO_2	2.9(-7)	4.6(-7)	7.4(-7)	2.3(-6)	1.6(-5)	5.5(-5)	9.3(-5)	1.2(-4)	1.3(-4)
(36)	O + O_2 + O_2	→	O_3 + O_2	9.8(-34)	1.1(-33)	1.1(-33)	1.1(-33)	1.0(-33)	9.5(-34)	8.2(-34)	7.4(-34)	6.7(-34)
(37)	O + O_2 + N_2	→	O_3 + N_2	1.0(-33)	1.2(-33)	1.2(-33)	1.1(-33)	1.1(-33)	1.0(-33)	8.7(-34)	7.8(-34)	7.4(-34)
(38)	O + O_3	→	O_2 + O_2	8.3(-16)	4.5(-16)	4.5(-16)	5.7(-16)	7.2(-16)	9.8(-16)	1.9(-15)	3.0(-15)	3.9(-15)
(43)	O + NO_2	→	O_2 + NO	9.1(-12)	9.1(-12)	9.1(-12)	9.1(-12)	9.1(-12)	9.1(-12)	9.1(-12)	9.1(-12)	9.1(-12)
(50)	O + HO_2	→	O_2 + OH	8.0(-11)	8.0(-11)	8.0(-11)	8.0(-11)	8.0(-11)	8.0(-11)	8.0(-11)	8.0(-11)	8.0(-11)
(71)	$O(^1D)$ + N_2O	→	NO + NO	1.1(-10)	1.1(-10)	1.1(-10)	1.1(-10)	1.1(-10)	1.1(-10)	1.1(-10)	1.1(-10)	1.1(-10)
(73)	$O(^1D)$ + H_2O	→	OH + OH	3.5(-10)	3.5(-10)	3.5(-10)	3.5(-10)	3.5(-10)	3.5(-10)	3.5(-10)	3.5(-10)	3.5(-10)
(94)	O_3 + NO	→	O_2 + NO_2	4.8(-15)	3.5(-15)	3.5(-15)	4.0(-15)	4.5(-15)	5.2(-15)	7.3(-15)	9.4(-15)	1.1(-14)
(95)	O_3 + NO	→	O_2 + NO_2^*	1.4(-16)	7.8(-17)	7.8(-17)	9.7(-17)	1.2(-16)	1.6(-16)	2.8(-16)	4.4(-16)	5.6(-16)
(101)	O_3 + OH	→	O_2 + HO_2	2.0(-14)	1.6(-14)	1.6(-14)	1.7(-14)	1.9(-14)	2.2(-14)	2.9(-14)	3.6(-14)	4.0(-14)
(115)	NO + NO_3	→	NO_2 + NO_2	8.7(-12)	8.7(-12)	8.7(-12)	8.7(-12)	8.7(-12)	8.7(-12)	8.7(-12)	8.7(-12)	8.7(-12)
(117)	NO + HO_2	→	OH + NO_2	2.0(-13)	2.0(-13)	2.0(-13)	2.0(-13)	2.0(-13)	2.0(-13)	2.0(-13)	2.0(-13)	2.0(-13)
(119)	NO + HO_2 + M	→	HNO_3 + M	3.3(-33)	3.3(-33)	3.3(-33)	3.3(-33)	3.3(-33)	3.3(-33)	3.3(-33)	3.3(-33)	3.3(-33)
(123)	OH + NO_2 + M	→	HNO_3 + M	4.5(-31)	4.5(-31)	4.5(-31)	4.5(-31)	4.5(-31)	4.5(-31)	4.5(-31)	4.5(-31)	4.5(-31)
(126)	H + O_2 + M	→	HO_2 + M	7.5(-32)	8.0(-32)	8.0(-32)	7.8(-32)	7.6(-32)	7.3(-32)	6.7(-32)	6.3(-32)	6.1(-32)
(132)	OH + HO_2	→	O_2 + H_2O	2.0(-11)	2.0(-11)	2.0(-11)	2.0(-11)	2.0(-11)	2.0(-11)	2.0(-11)	2.0(-11)	2.0(-11)
(134)	OH + CO	→	H + CO_2	1.4(-13)	1.4(-13)	1.4(-13)	1.4(-13)	1.4(-13)	1.4(-13)	1.4(-13)	1.4(-13)	1.4(-13)
(135)	OH + CH_4	→	H_2O + CH_3	1.3(-15)	8.2(-16)	8.2(-16)	9.8(-16)	1.2(-15)	1.5(-15)	2.4(-15)	3.5(-15)	4.3(-15)

*Rate constants for photolytic reactions and for two-body reactions are given in cm^3sec^{-1} and for three-body reactions units are cm^6sec^{-1}.

For nitric acid in Figs. 1h and 2h, the calculated values above 20 km agree quite well with measurements of Evans et al. [10]; but at 20 km and below, the measurements of Evans et al. and of Lazrus and Gandrud [11] are somewhat lower than the calculated values. A shift in the area of peak concentration between measured and calculated values is also observed.

Calculated values of nitrous oxide in Figs. 1i and 2i are shown to agree with measurements taken by Schmeltekopf et al. [12] in early spring (May 1976) and not quite as well with measurements taken in late summer (August 1975). Measurements in May of 1976 were taken in Alaska, and measurements in August of 1975 were taken at Saskatchewan.

Although there is some information in the literature for experimental measurements of other species in the stratosphere such as CH_4, CO, H_2O, CO_2, and $O_2(^1D)$, no measurements were plotted on the graphs of calculated profiles for these species. However, differences in the calculated profiles of H_2O, CO, and CH_4 between 32° N [2] and 65° N seem to agree with comments on the latitudinal variation of these species by Farmer [13].

The calculated values of water vapor are well within the envelope presented from measurements by Mastenbrook in [14] and are extremely close to the average profile shown by Mastenbrook.

The authors do not intend to slight measurements performed by experimenters other than those mentioned in this report. Some measurements were not included simply to avoid crowding the presented graphs to such a degree as to make them unreadable. The intent is merely to compare some of the calculated results with some observations to show that the calculations are somewhat reasonable and to establish credibility in the modeling effort.

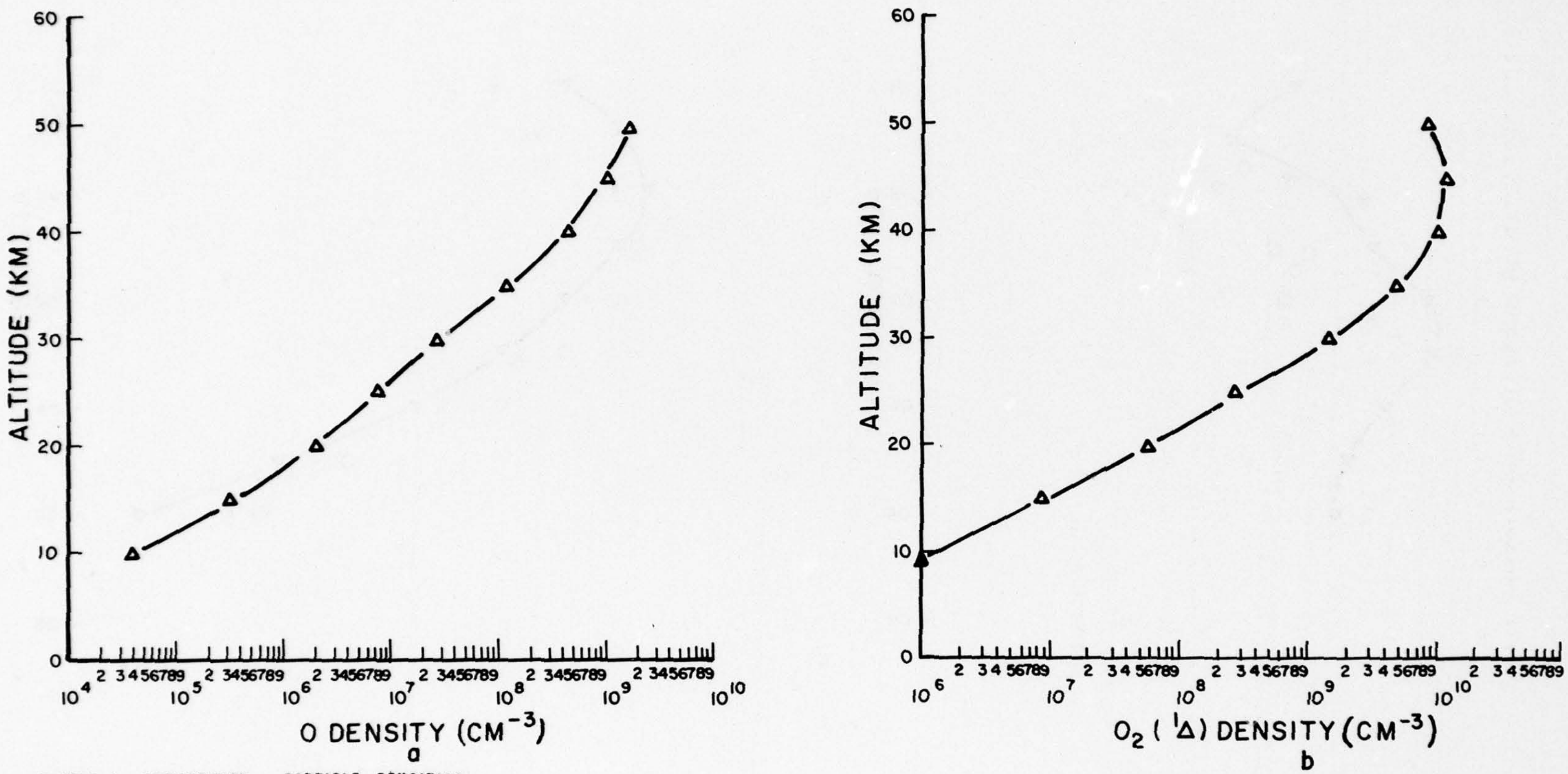
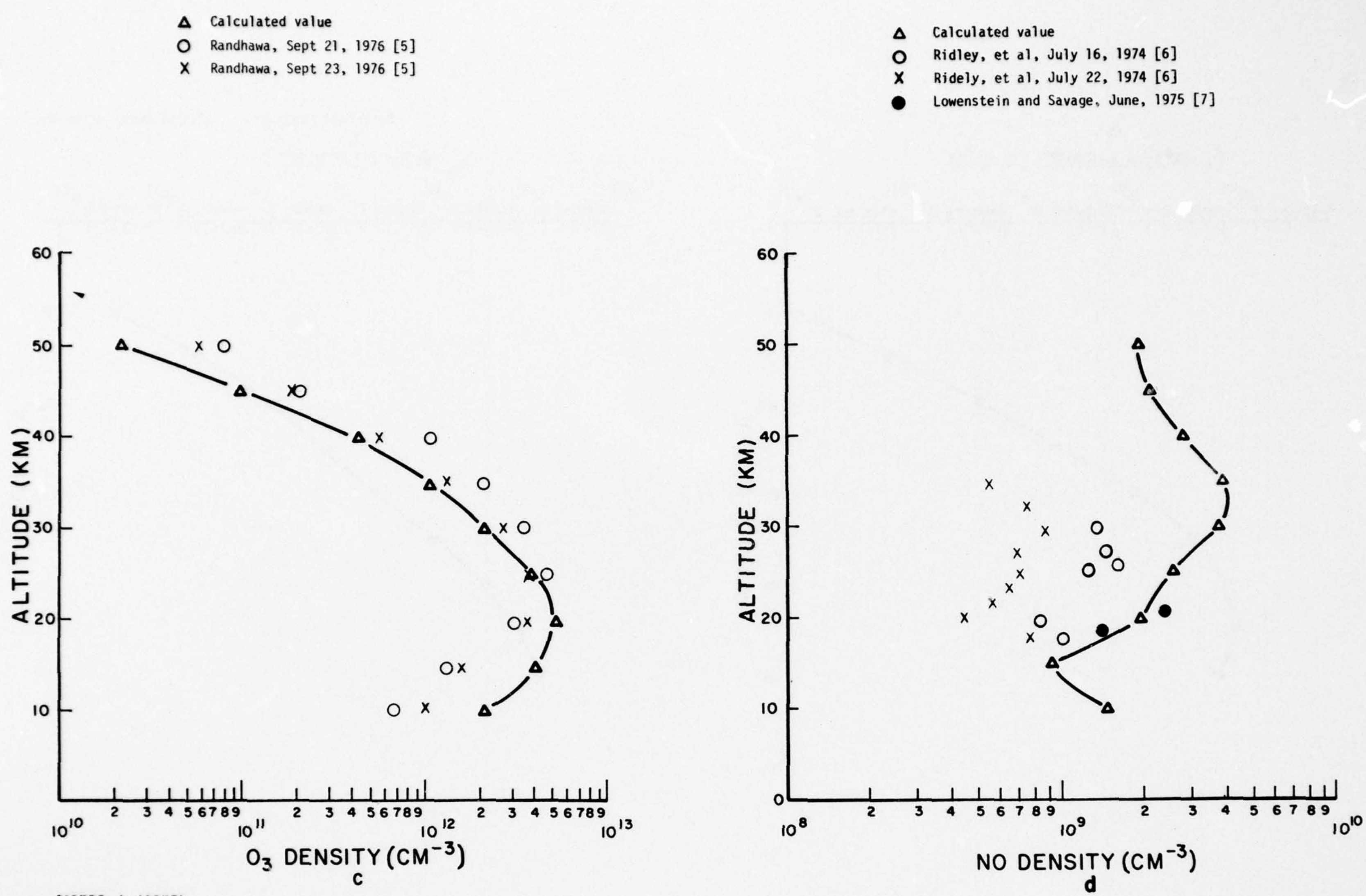


FIGURE 1. COMPOSITION - PARTICLE DENSITIES



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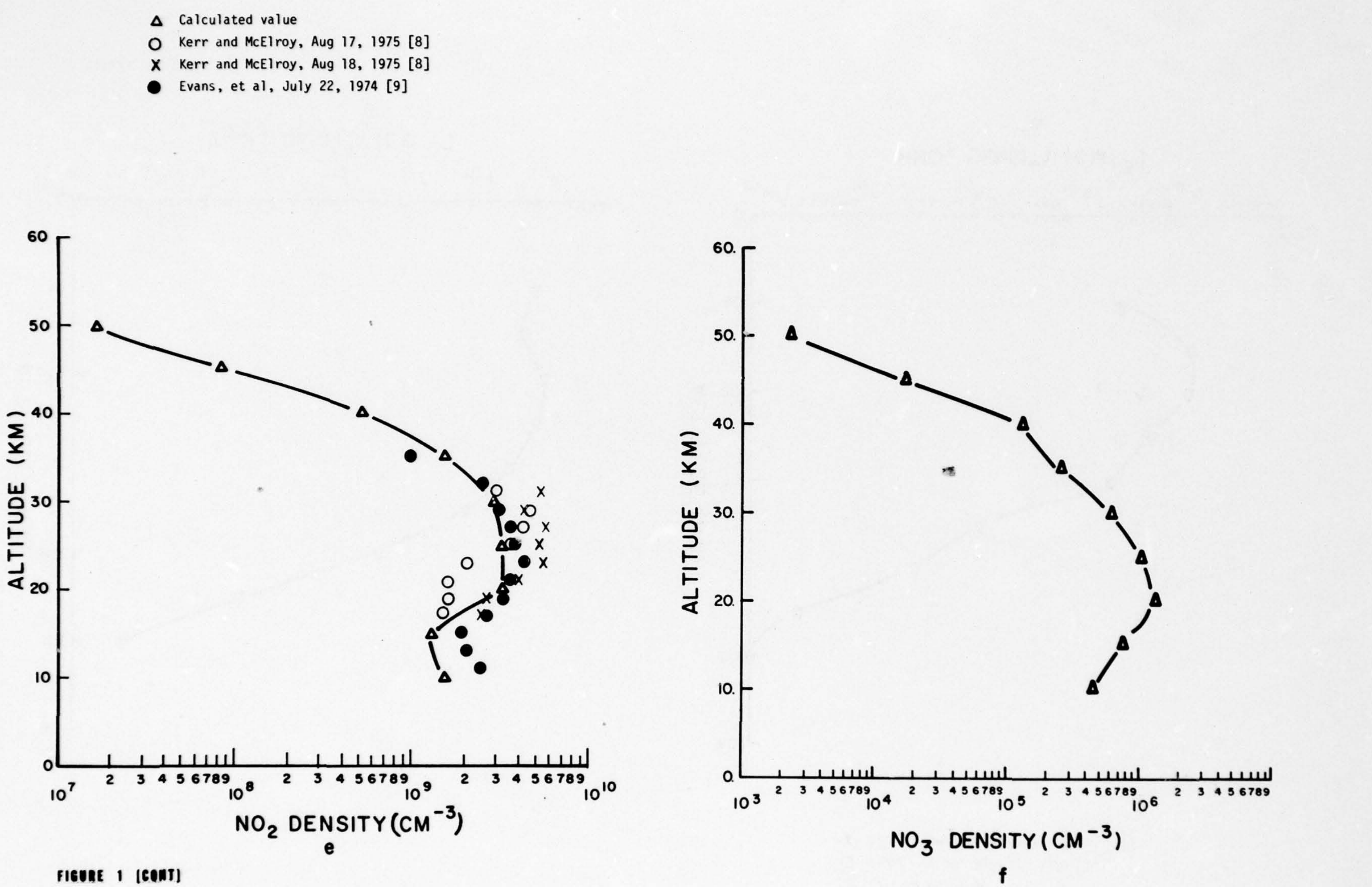

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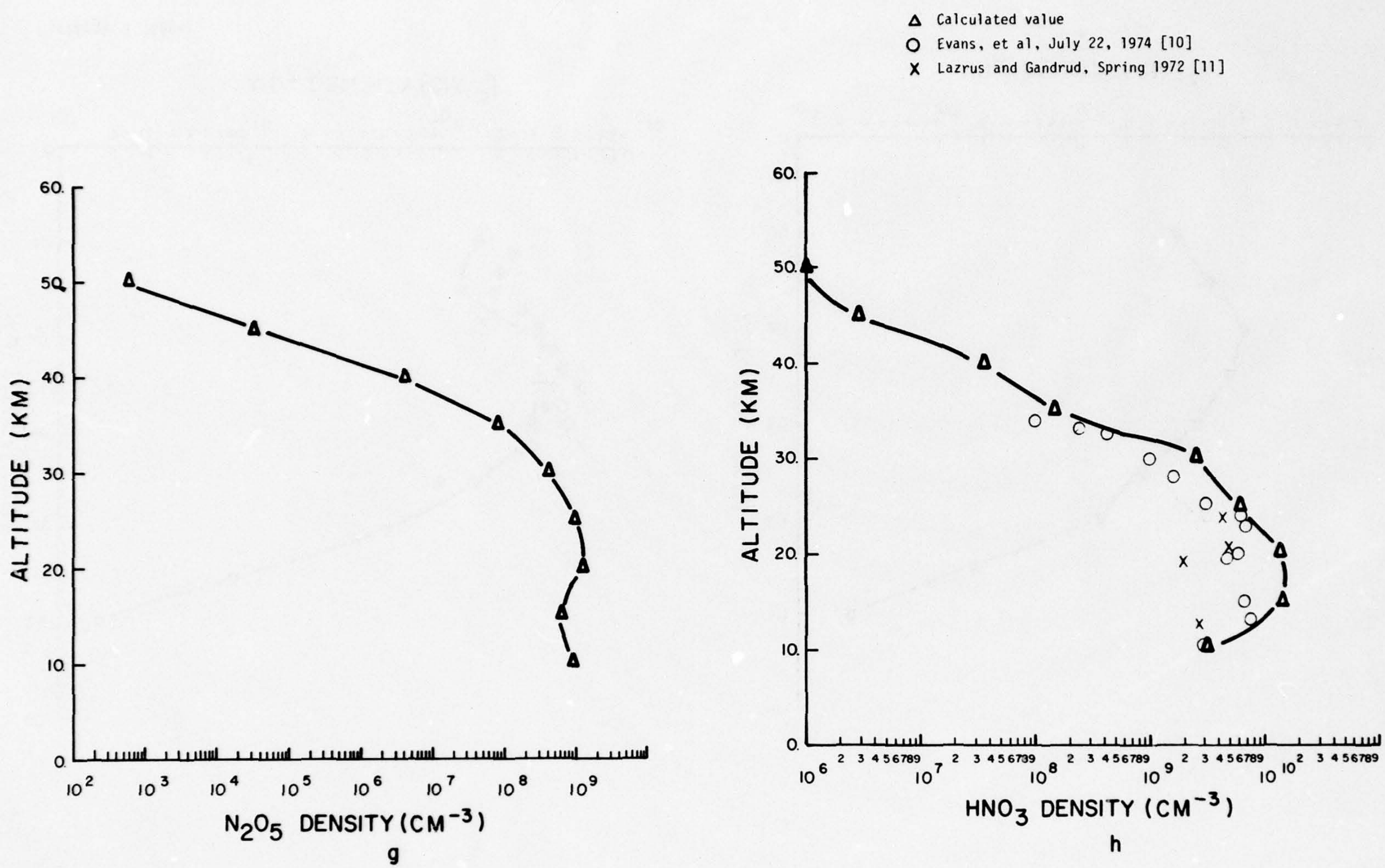

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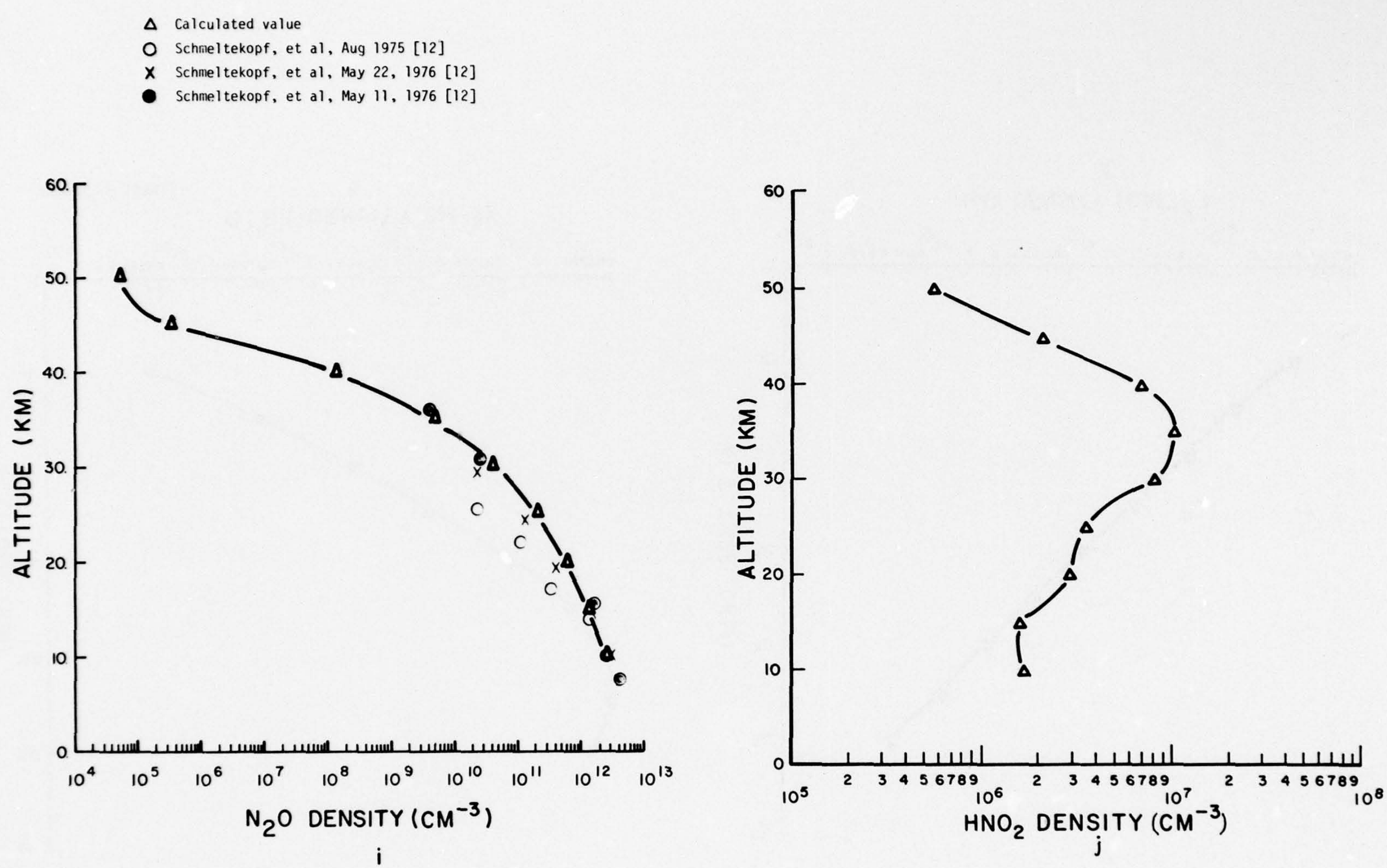

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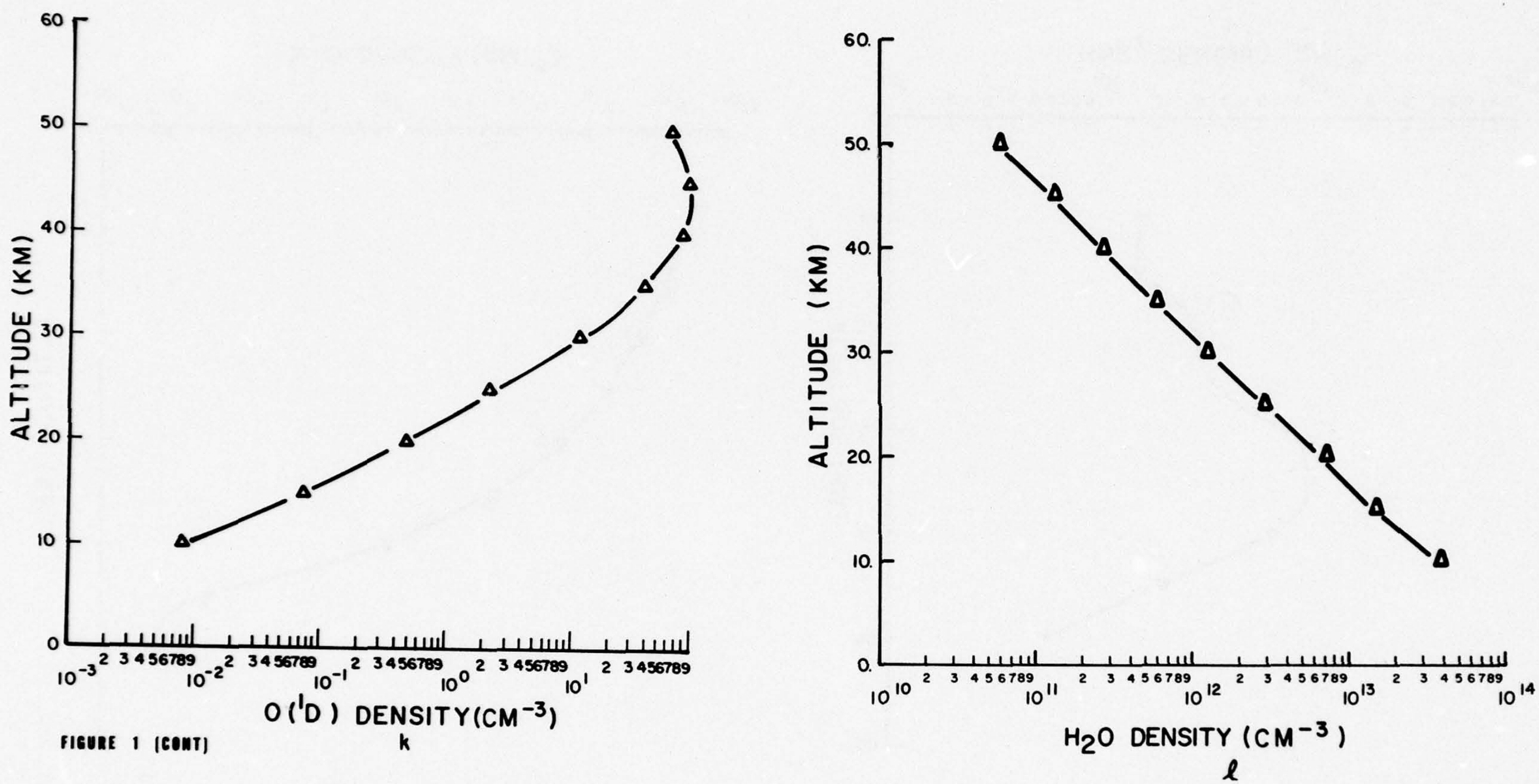

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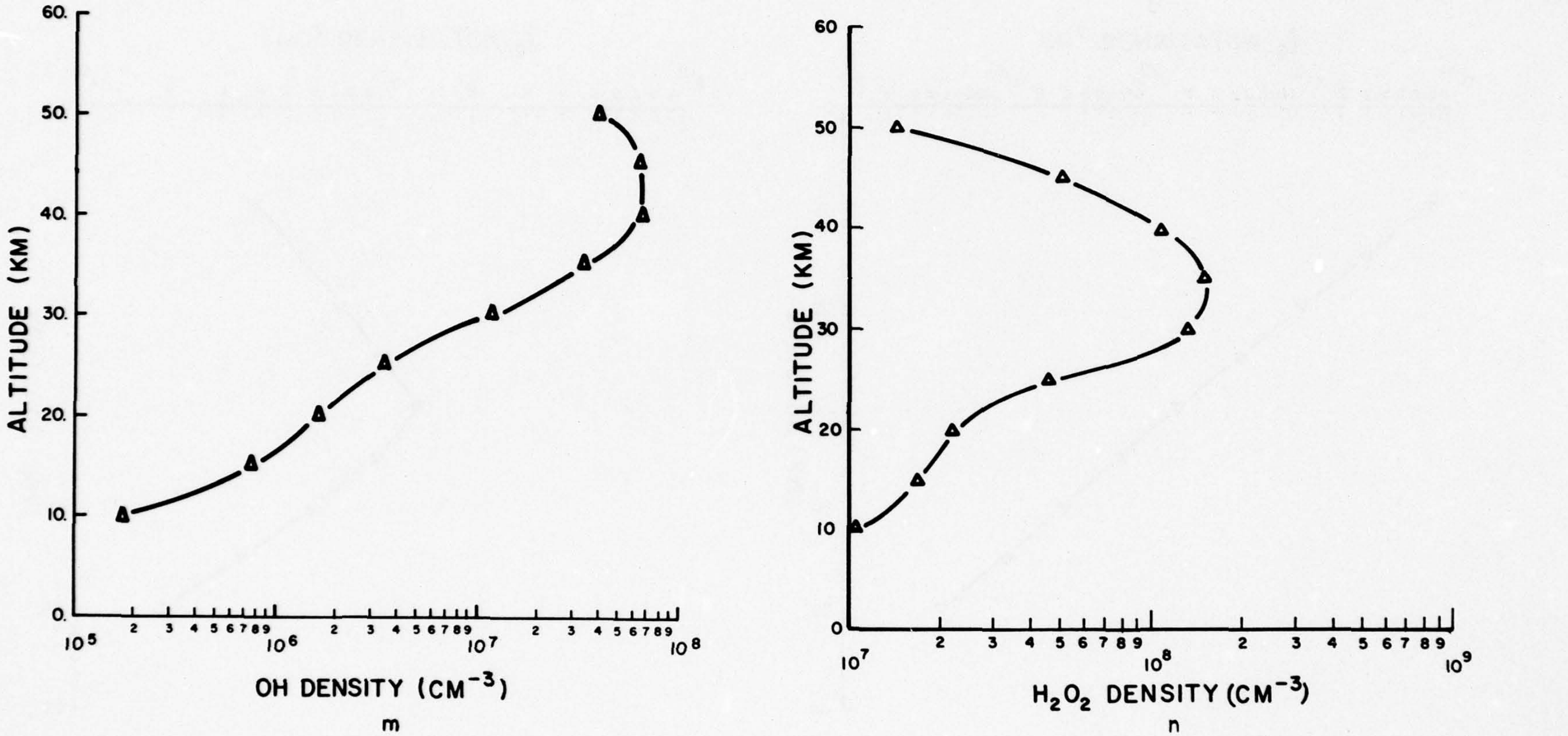

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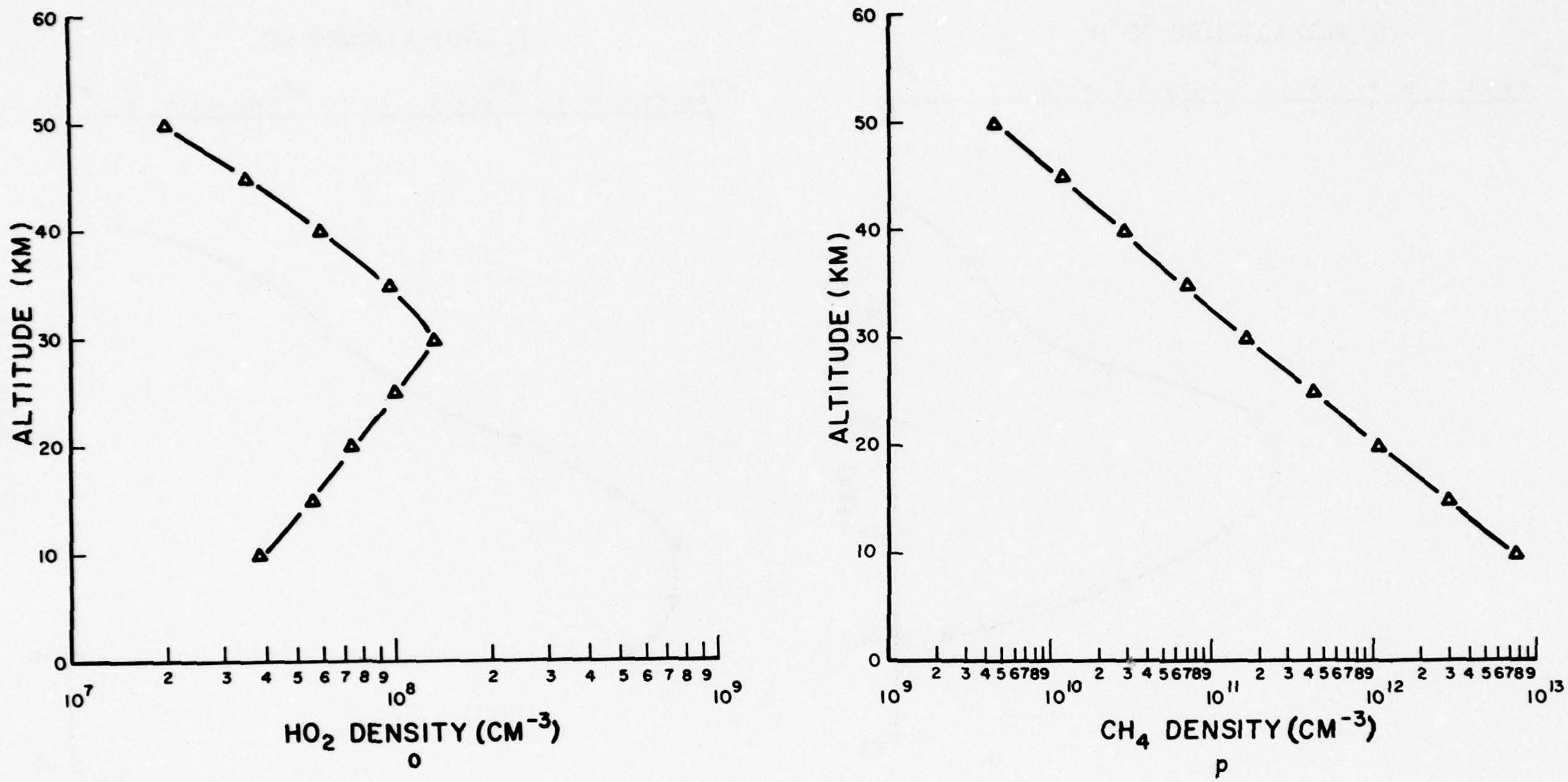

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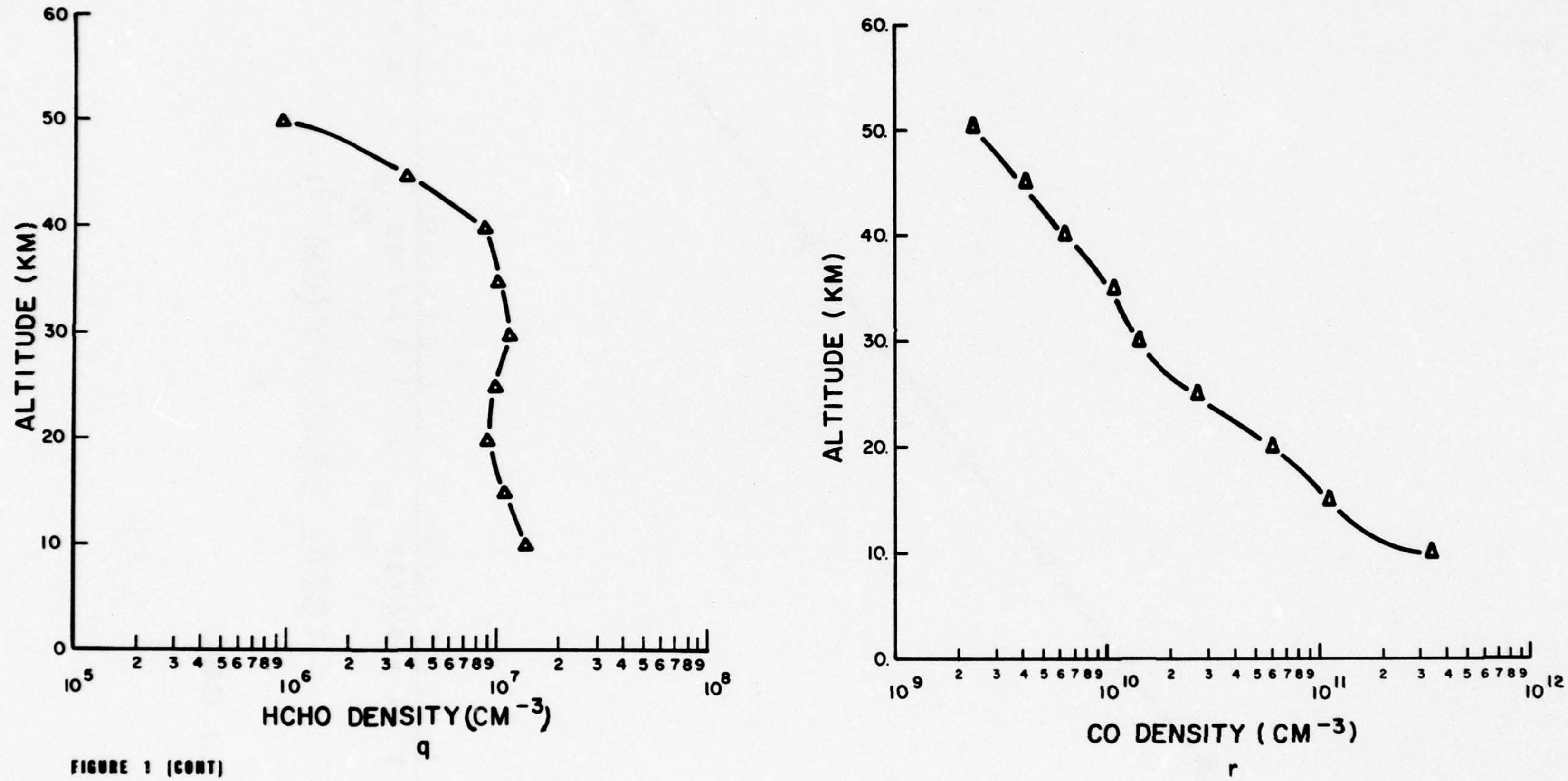

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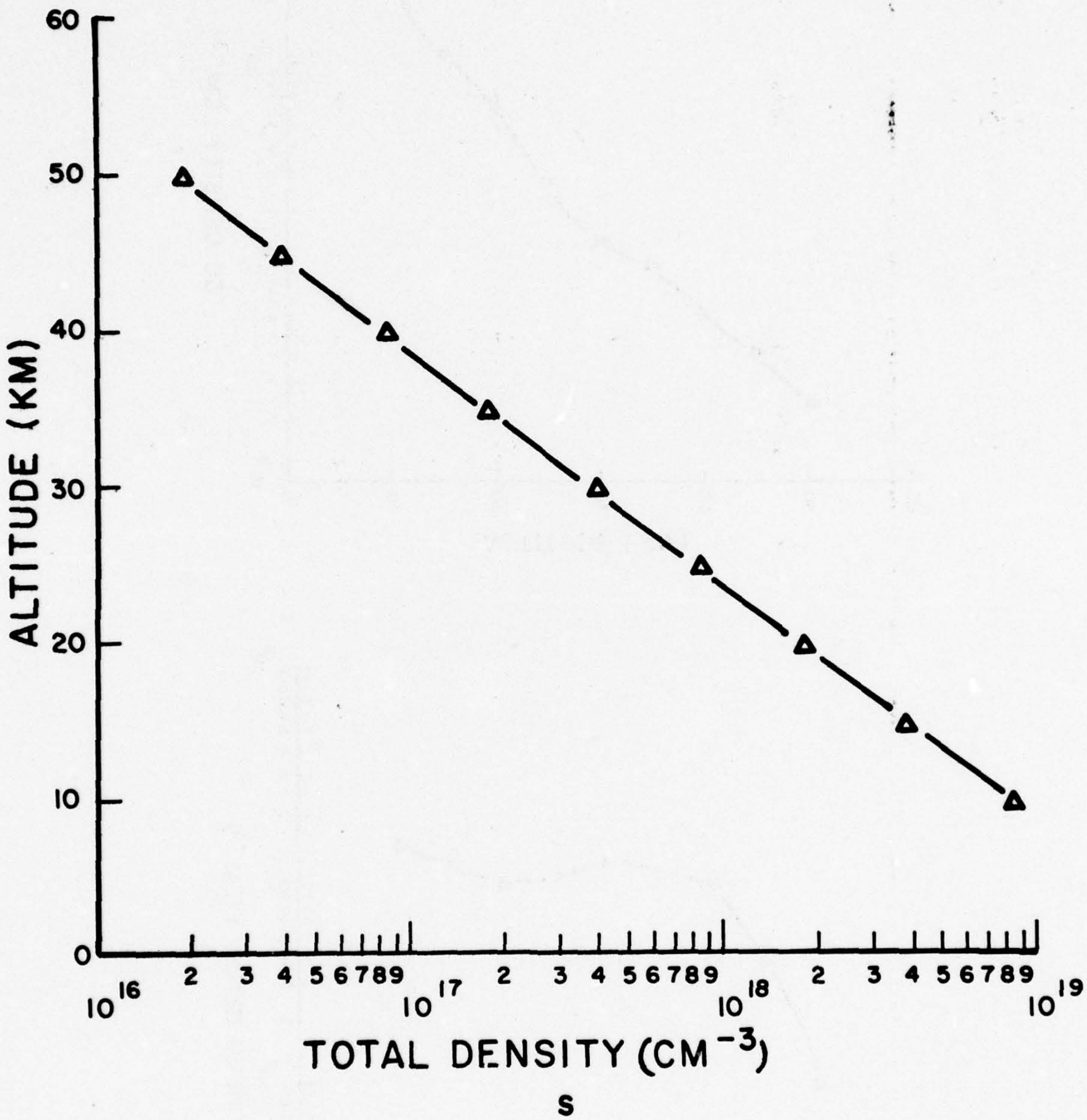

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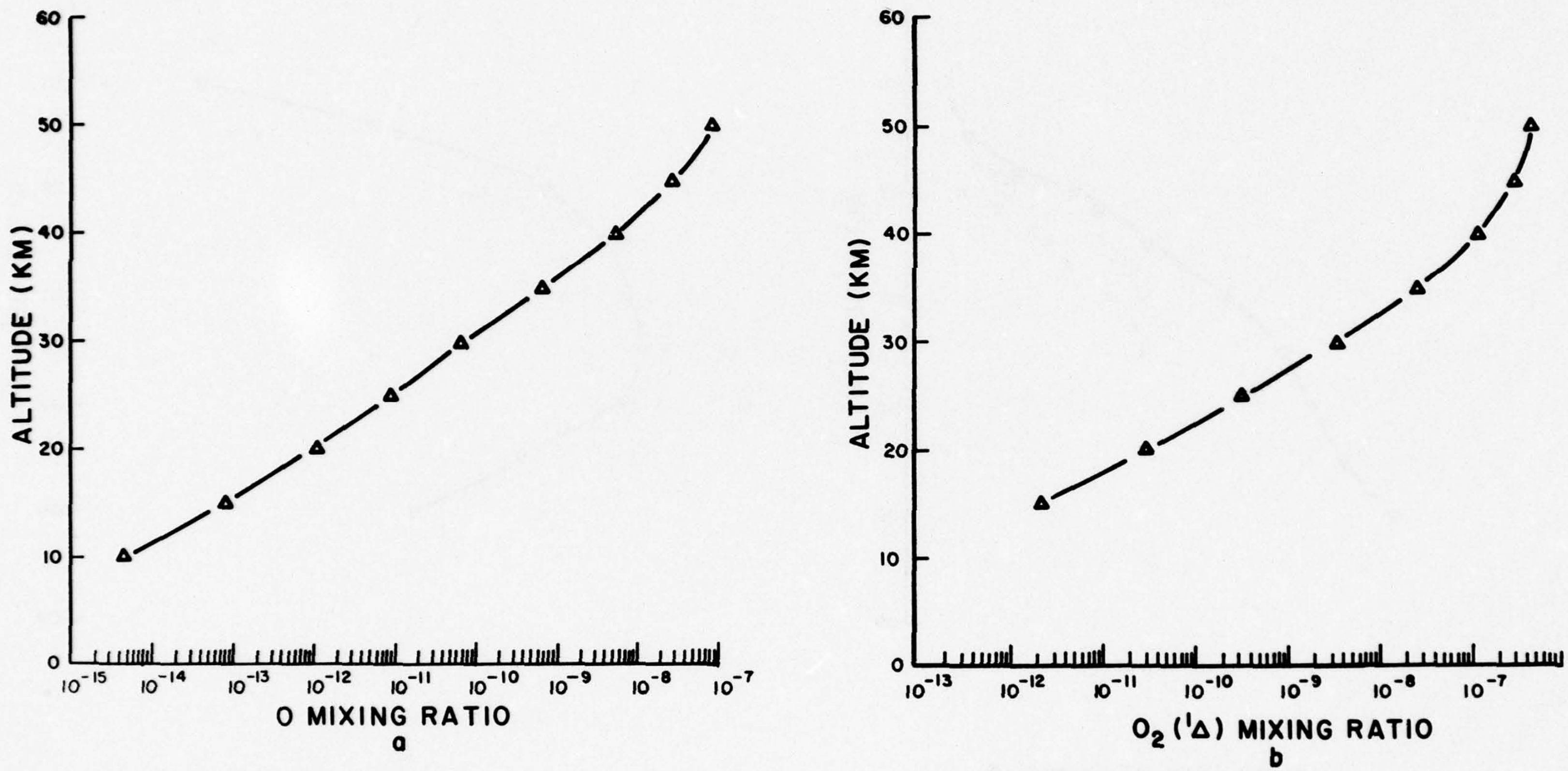
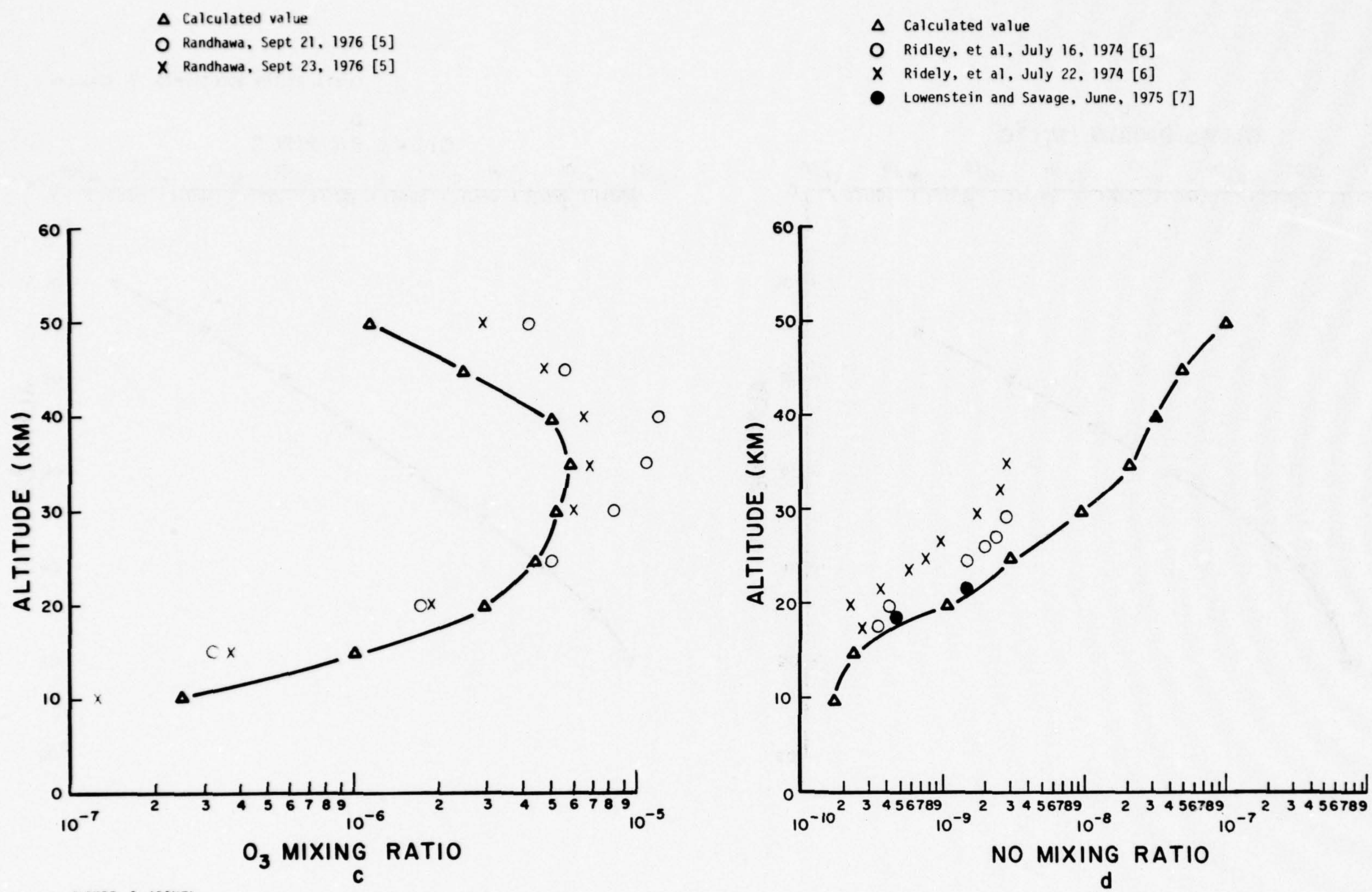


FIGURE 2 COMPOSITION MIXING RATIOS



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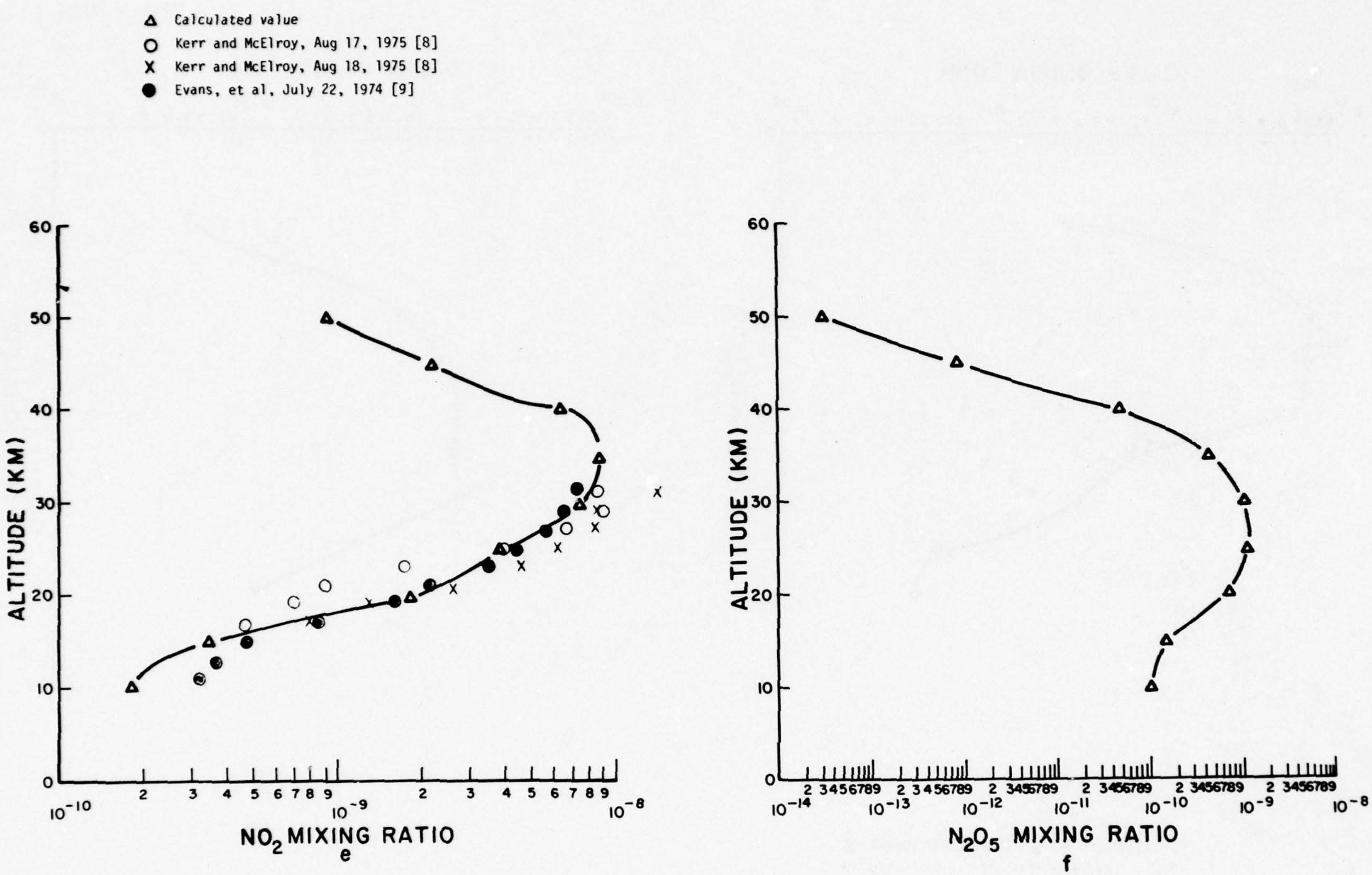

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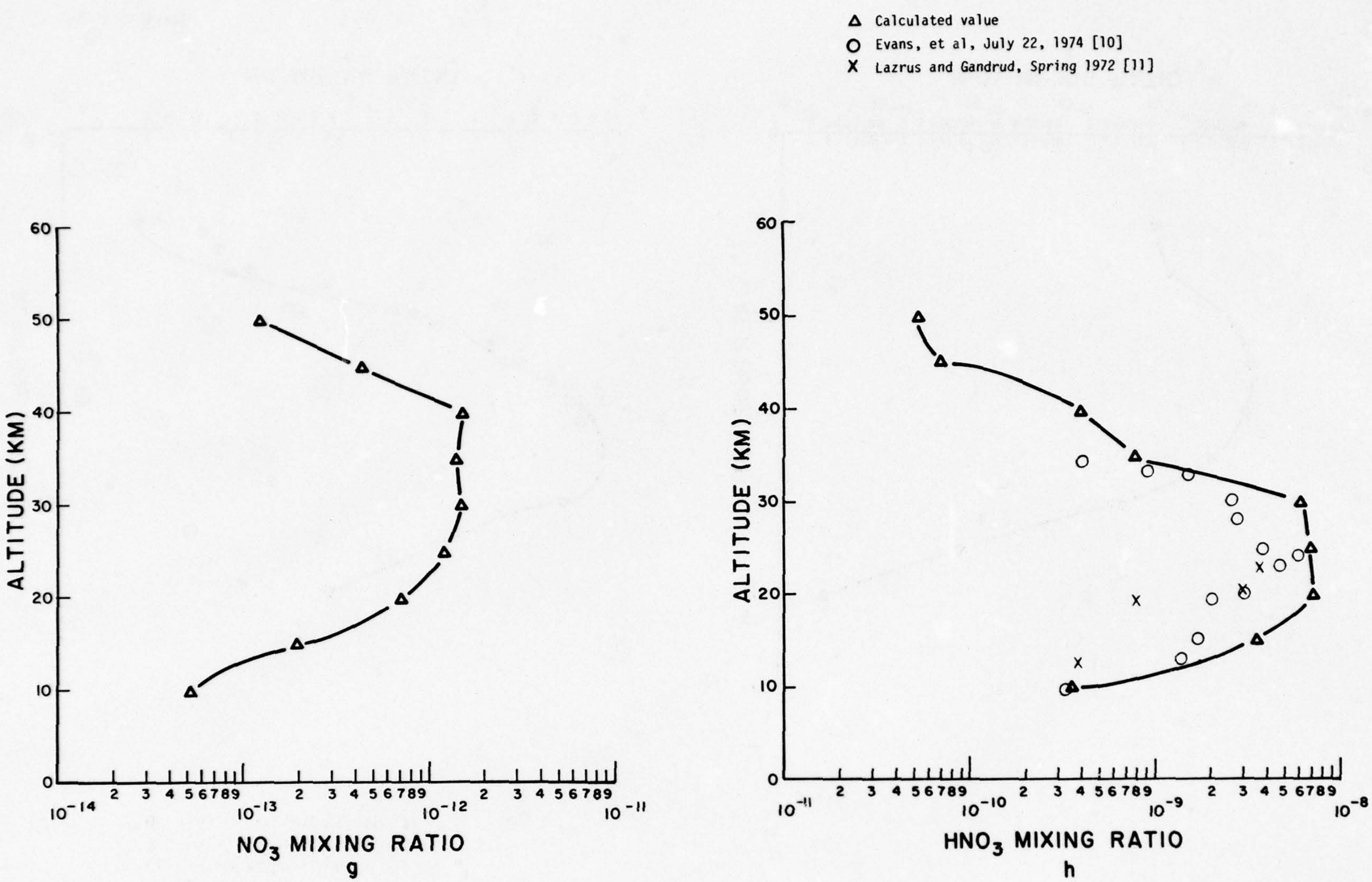

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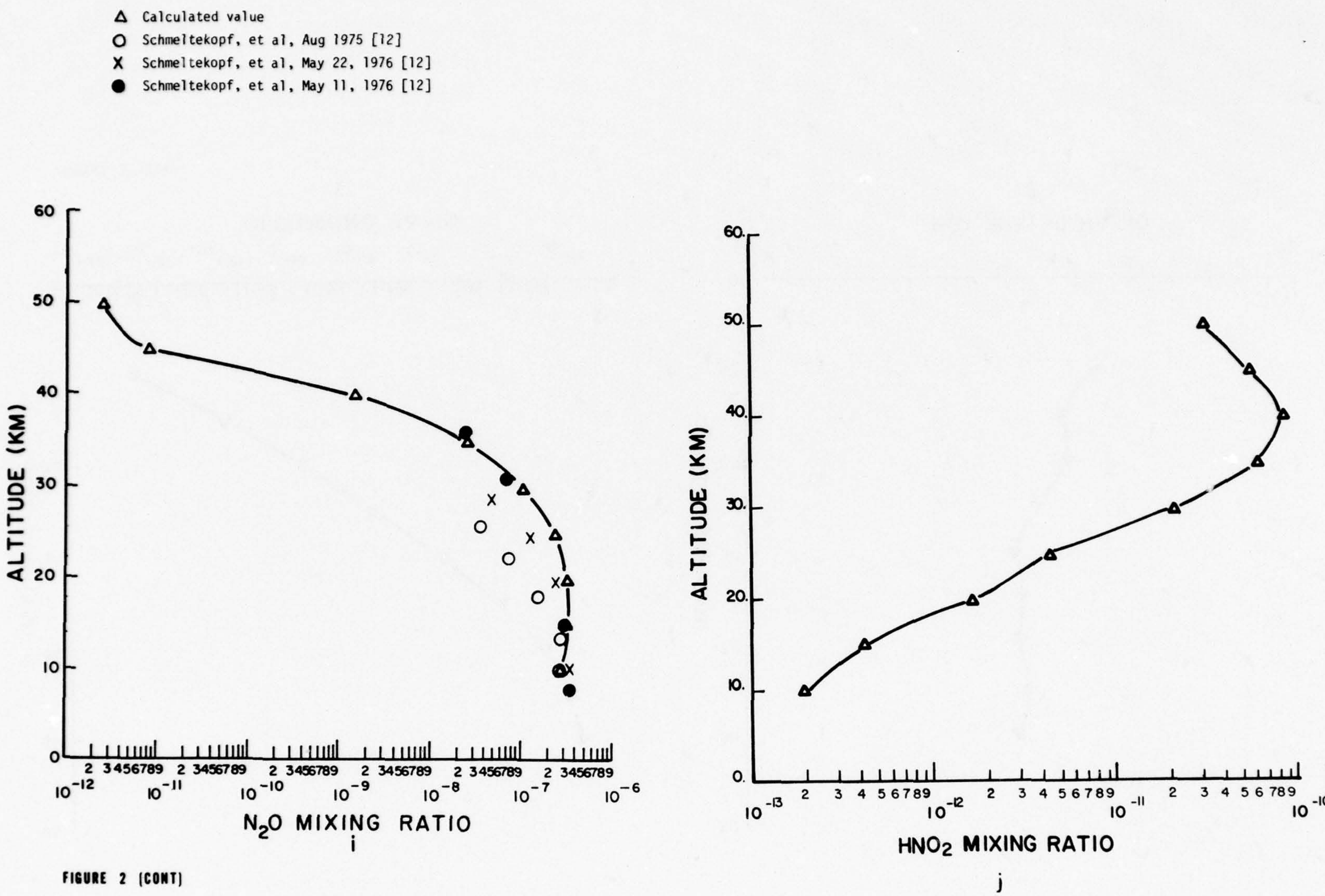

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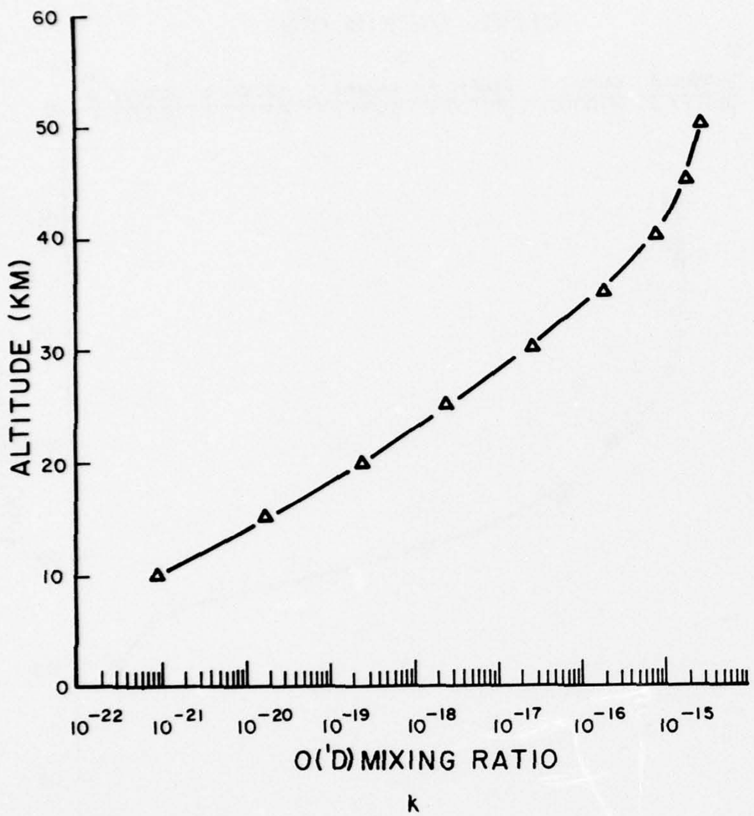

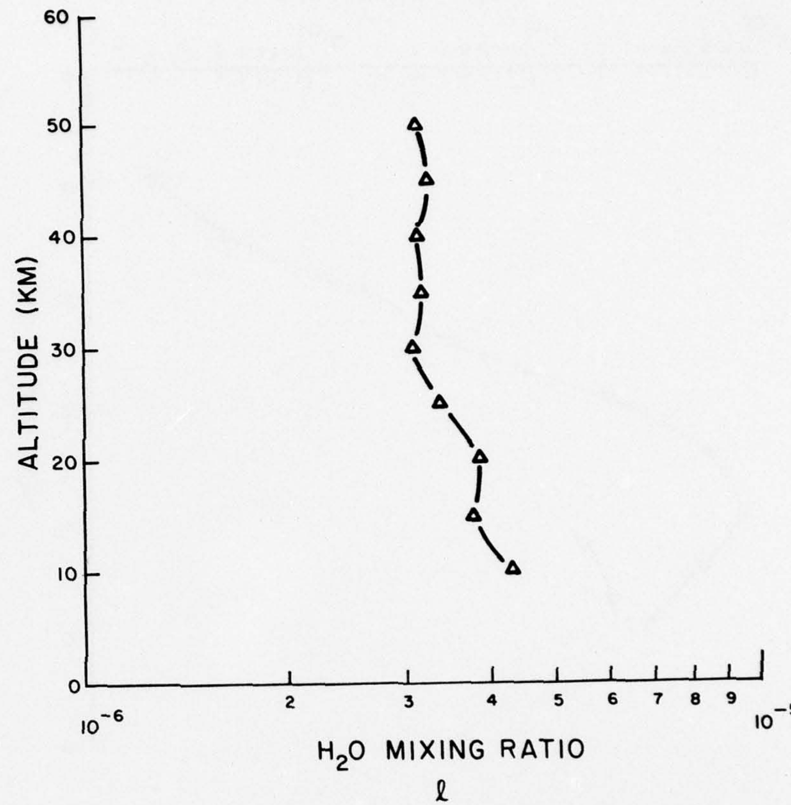

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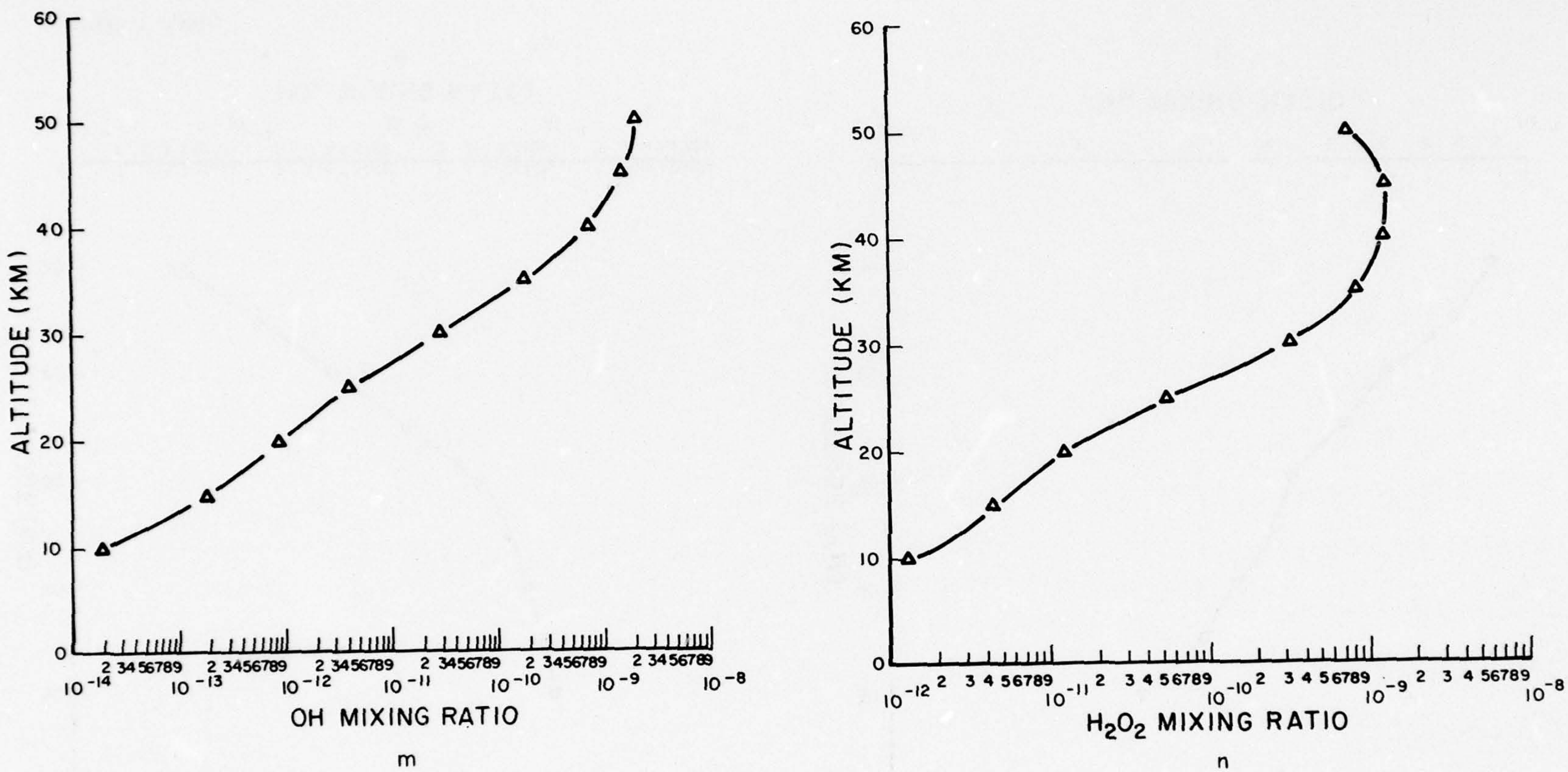

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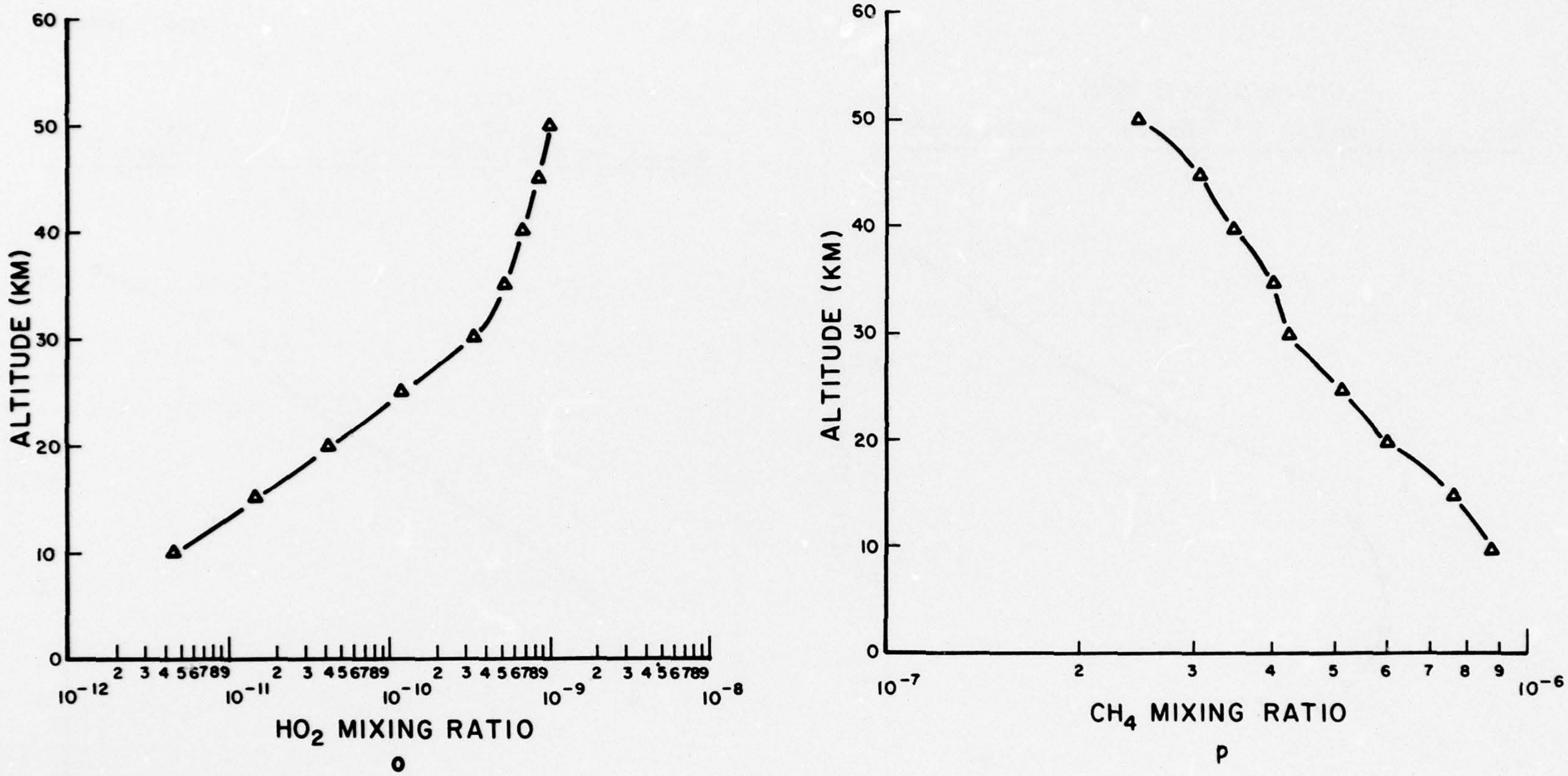

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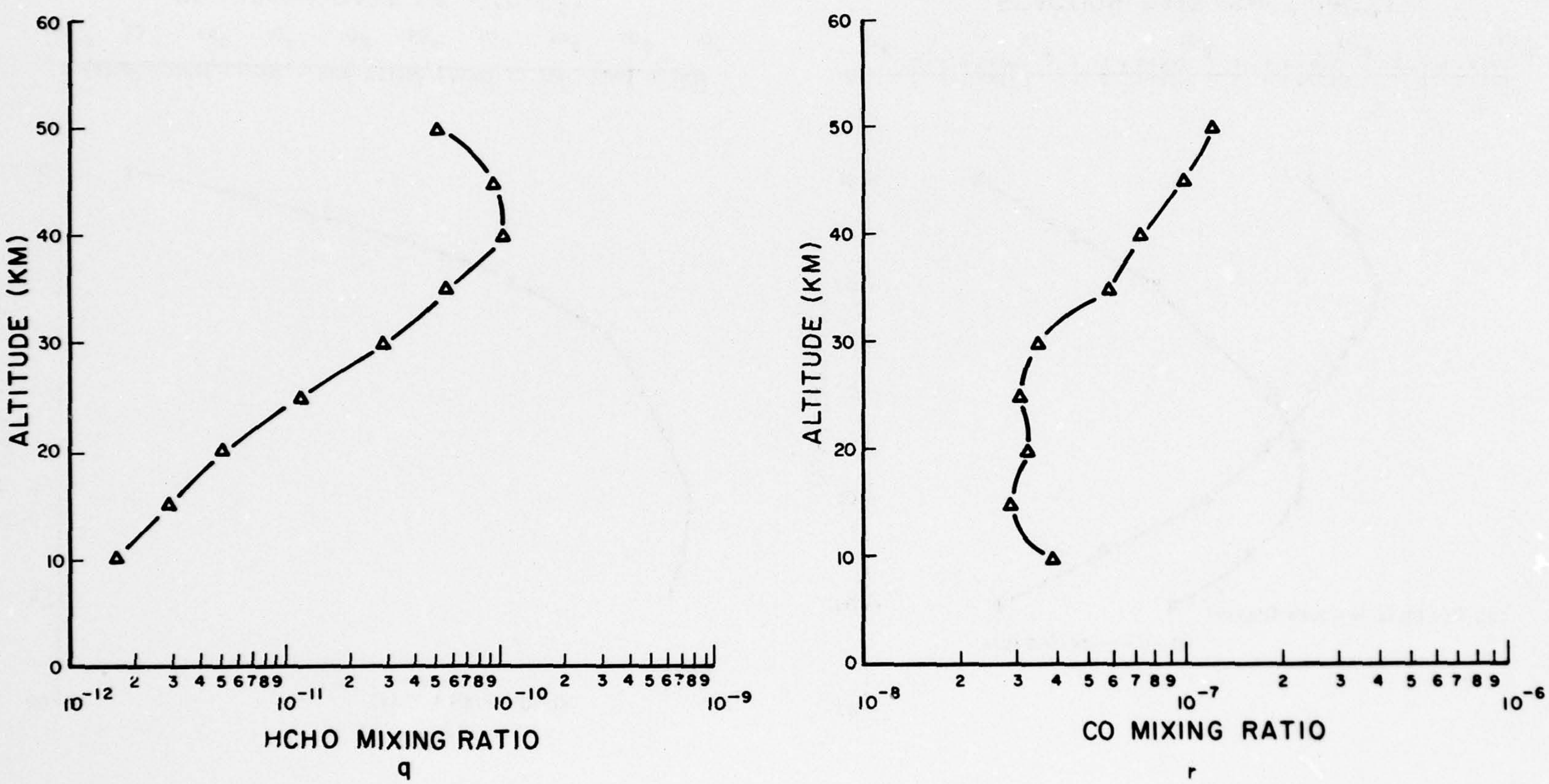

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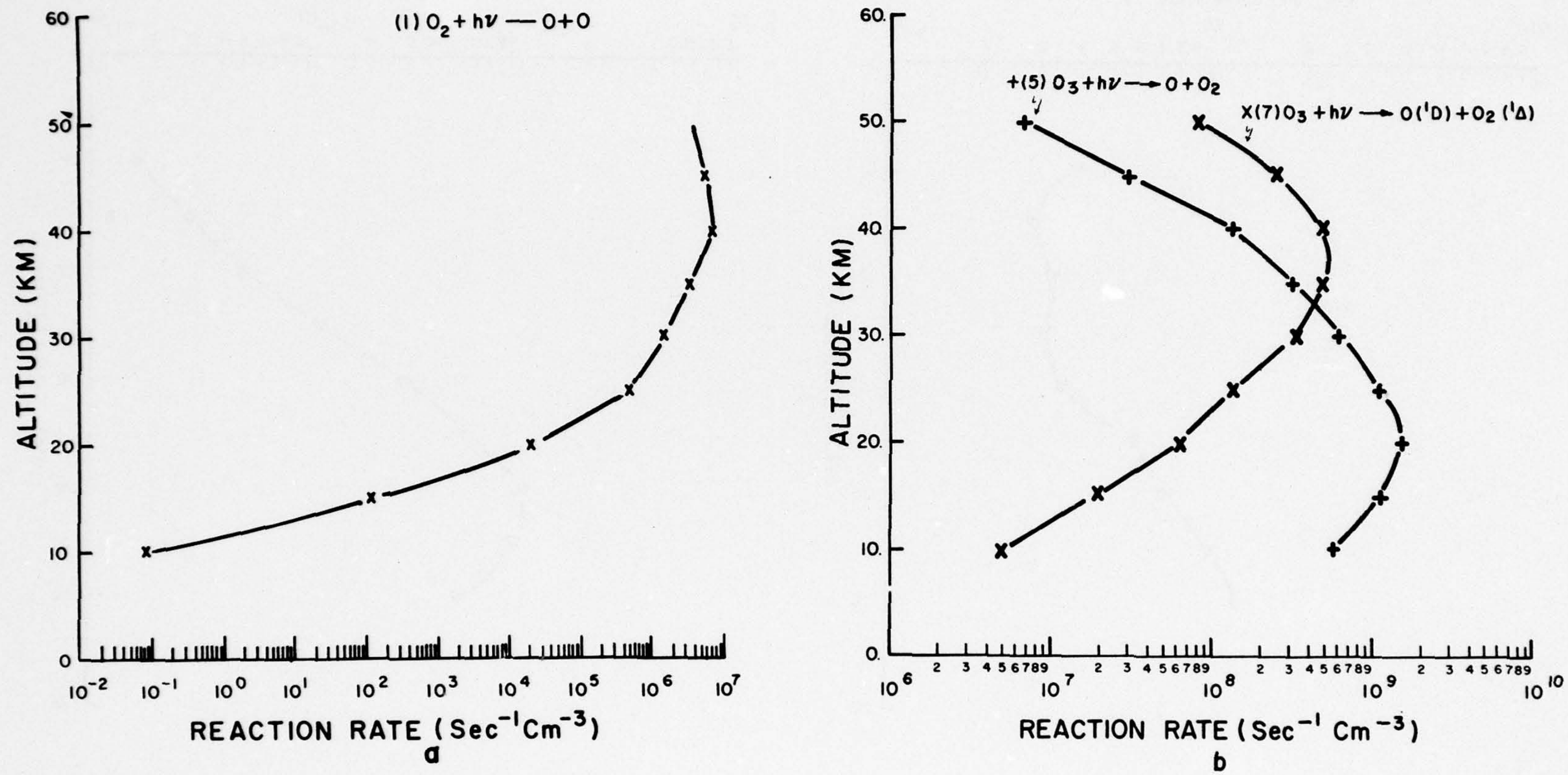
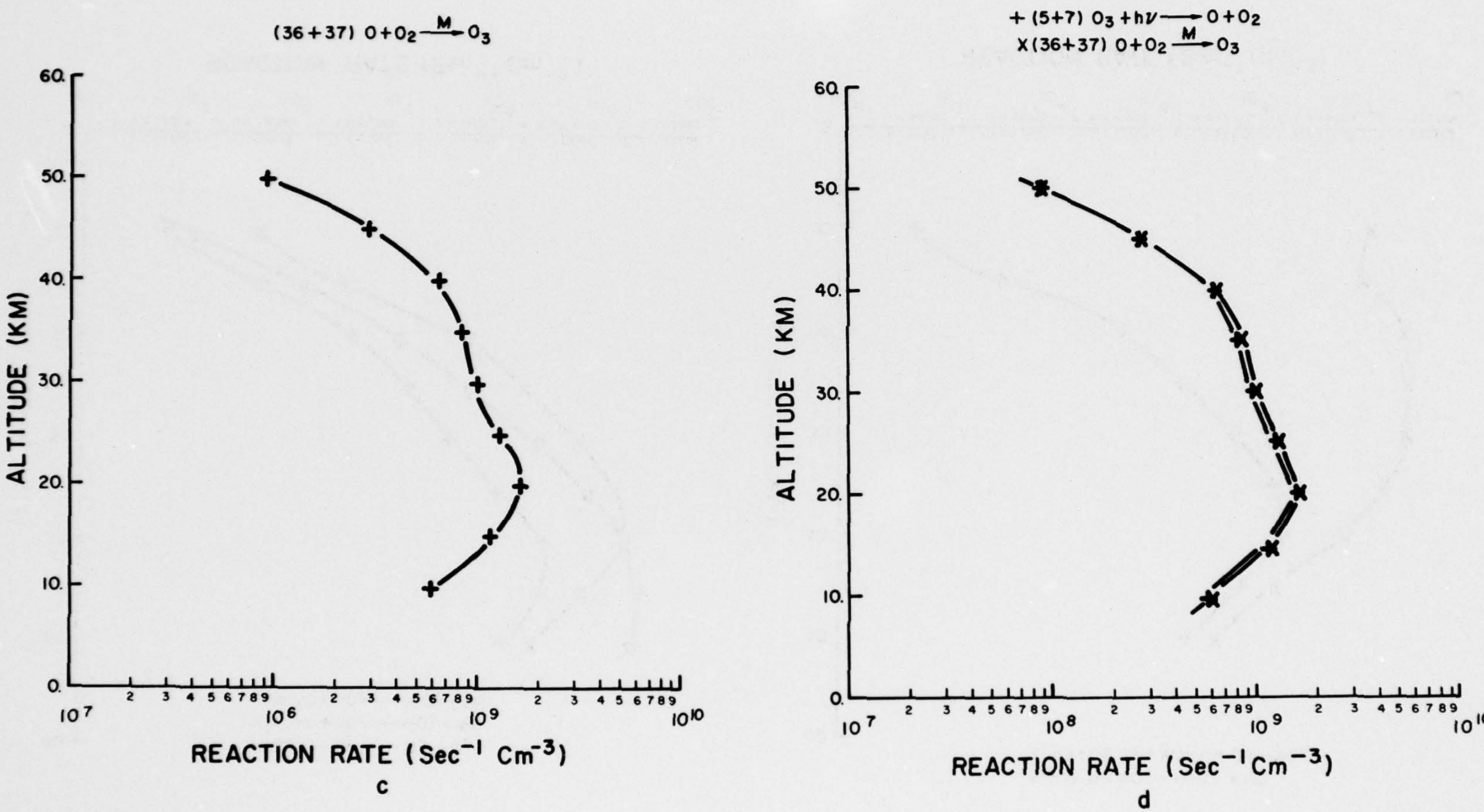


FIGURE 3. RATES OF CHEMICAL AND PHOTODISSOCIATION REACTIONS



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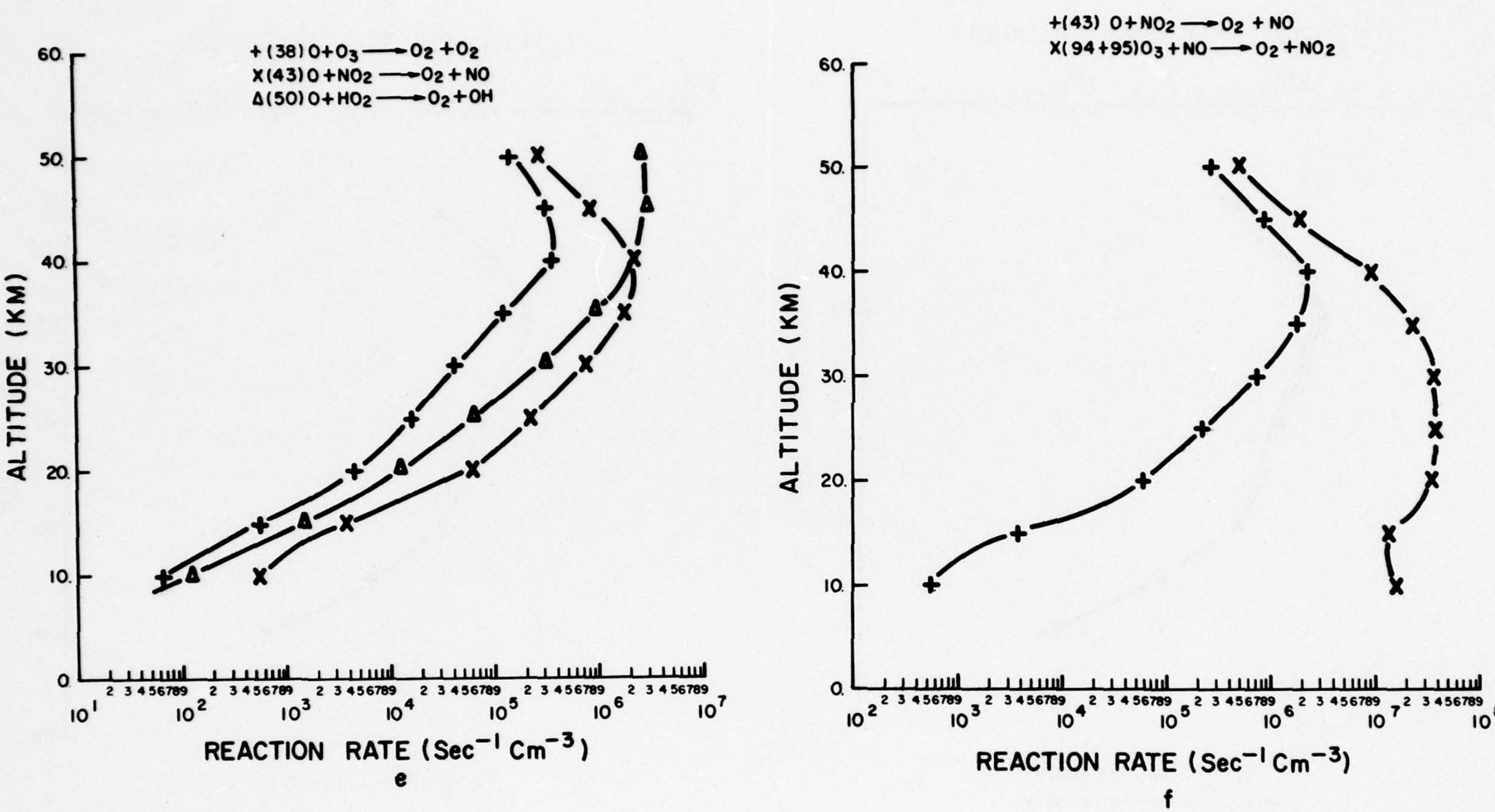

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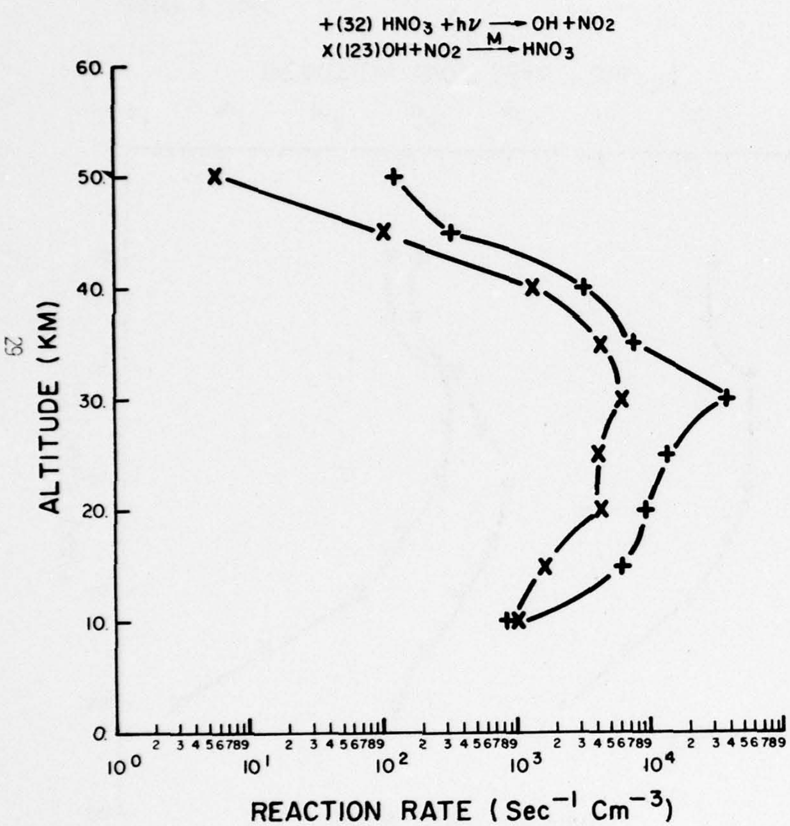

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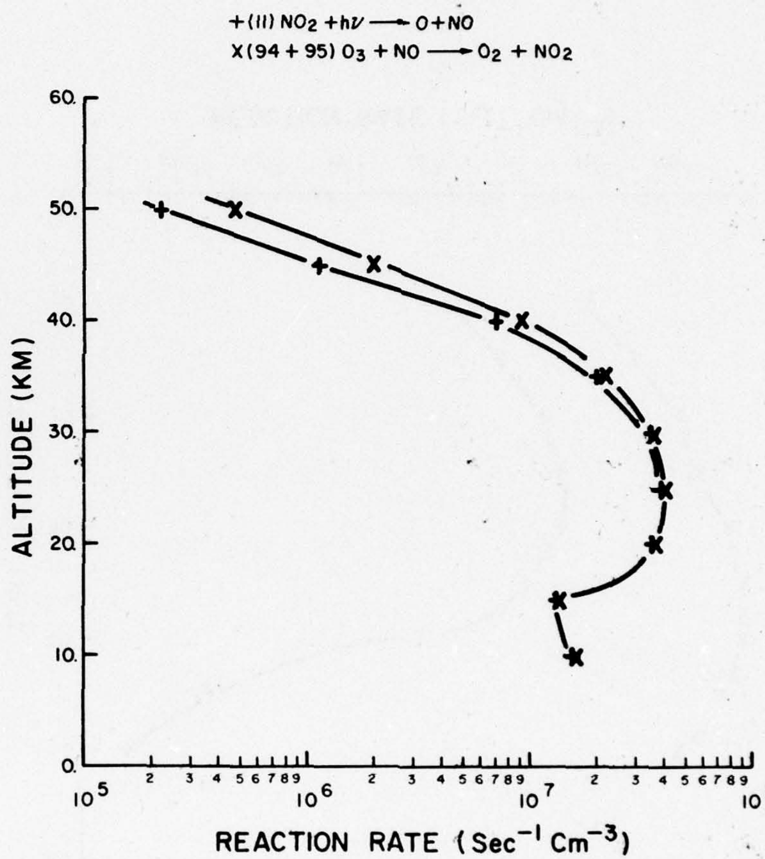

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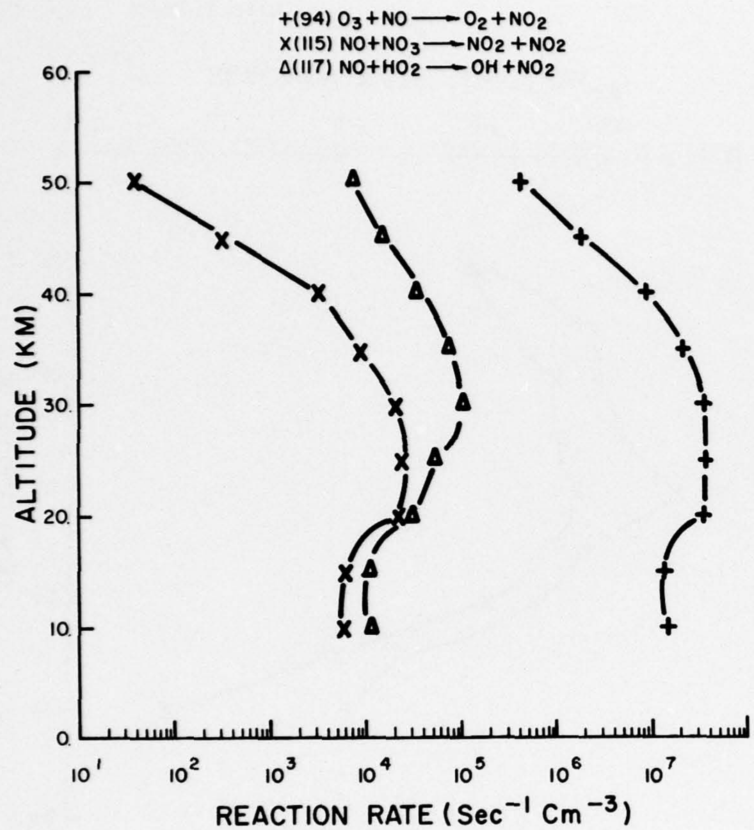
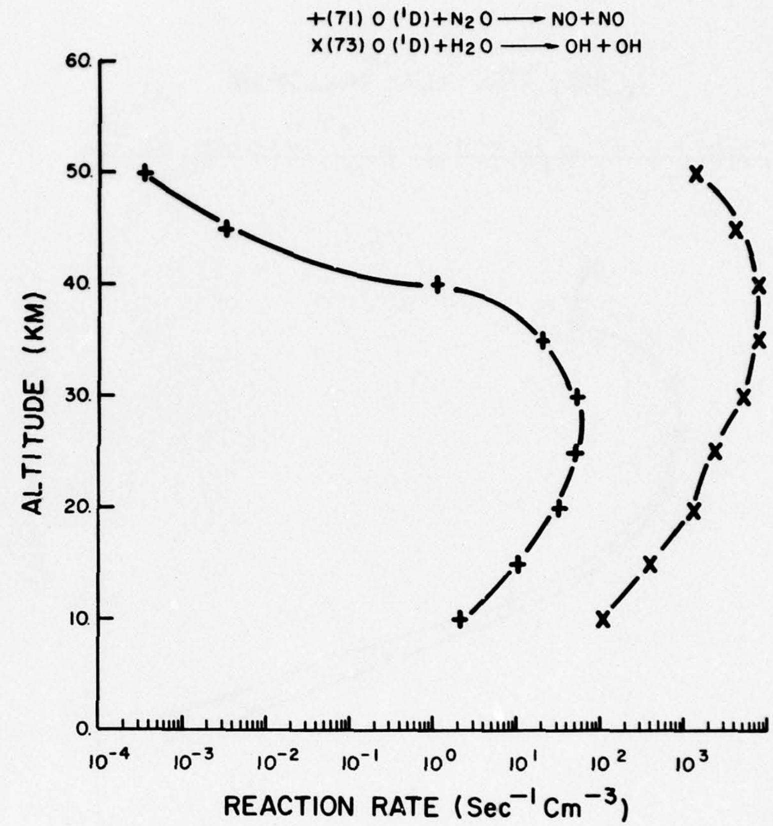
FIGURE 3 (CONT)



i



j

FIGURE 3 (CONT)

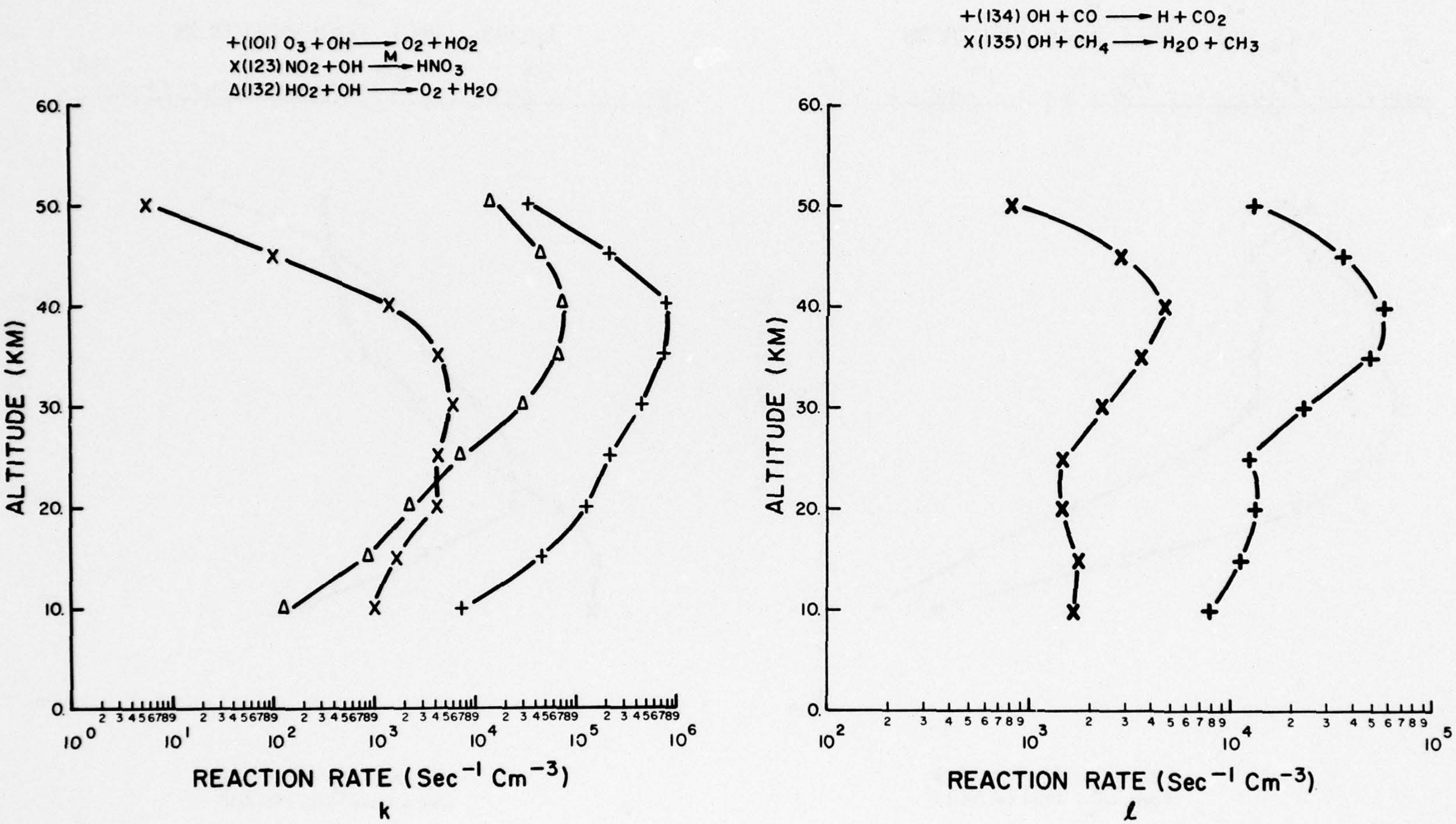


FIGURE 3 (CONT)

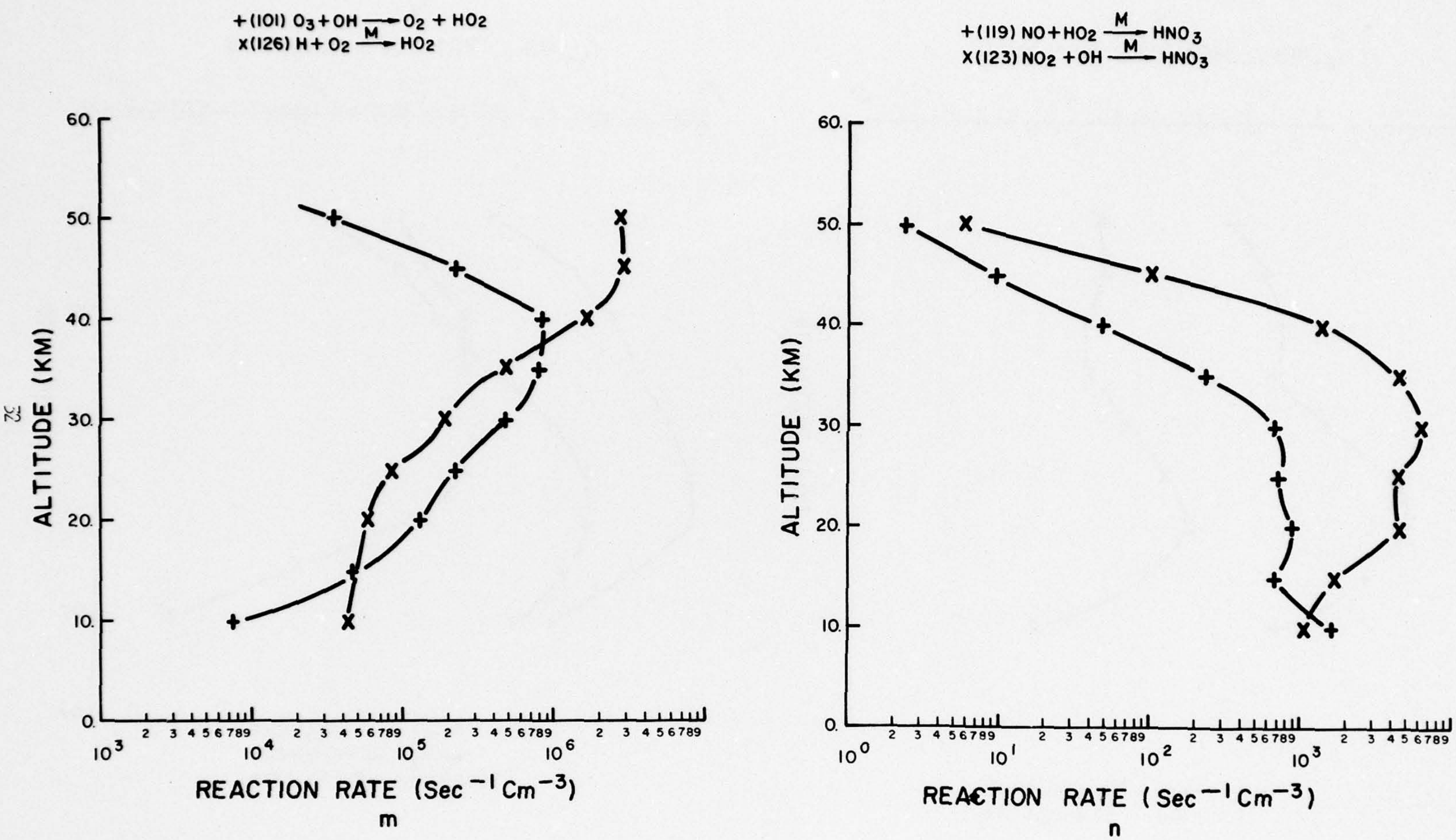


FIGURE 3 (CONT)

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